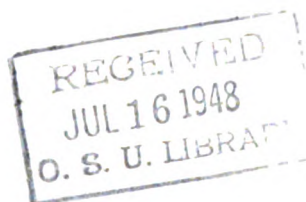


UNITED STATES
DEPARTMENT OF THE INTERIOR
J. A. KRUG, SECRETARY

BUREAU OF MINES
JAMES BOYD, DIRECTOR

INFORMATION CIRCULAR



SOME OBSERVATIONS ON COKING PRACTICE IN GERMANY

PART I. METALLURGICAL COKE

PART II. SLANTING TYPE DIDIER COKE OVENS,
STADTISCHE WERKE KARLSRUHE



BY

FRANK H. REED

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SOME OBSERVATIONS ON COKING PRACTICE IN GERMANY^{1/}

By Frank H. Reed^{2/}

FOREWORD

This report is one of a series written by members of the Solid Fuels Mission to Germany describing wartime developments in the mining, preparation, and utilization of coal. This mission was organized early in 1945 under the auspices of the Technical Industrial Intelligence Committee, which cooperated with a counterpart British committee. The Solid Fuels Mission operated under the direction of a steering committee headed by Dr. C. J. Potter, Deputy Solid Fuels Administrator. The personnel of the mission consisted of H. F. Yancey, in charge, J. W. Buch, Thomas Fraser, L. L. Newman, and L. D. Schmidt of the Bureau of Mines; H. H. Lowry, Director of the Coal Research Laboratory, Carnegie Institute of Technology; H. J. Rose, Vice-president and Director of Research, Bituminous Coal Research, Inc.; and F. H. Reed, Chief Chemist, Illinois State Geological Survey.

Over 50 reports were written by the members of the Solid Fuels Mission. These, together with microfilms of supporting data obtained in the course of the investigations, are on file at the Bureau of Mines, Washington, D. C., where they are available for public examination. The Office of the Publication Board in the Department of Commerce is prepared to furnish photolith copies, photostats, or microfilm reproduction of all declassified reports. A Bibliography of Scientific and Industrial Reports is issued weekly by the Department of Commerce, Office of the Publication Board, and is sold by the Superintendent of Documents, Government Printing Office, Washington 25, D. C., on a subscription basis. An initial payment of \$10 will provide for approximately 9 months, and subscribers will be notified when additional remittance is necessary. All subscriptions will begin with volume 1, No. 1, unless ordered otherwise. The Bibliography covers all materials and subjects and gives instructions for obtaining the desired items.

The reports of the Solid Fuels Mission relate to the following general subjects: Mining methods in the Ruhr district of Germany, with special reference to mechanical cutting and loading equipment, pneumatic packing, and Diesel mine locomotives; activities of industrial research laboratories supported by the German coal and coke industry; coal preparation in western Europe; low-temperature carbonization processes; metallurgical coke from

^{1/} The Bureau of Mines will welcome reprinting of this paper, provided the following footnote acknowledgment is used: "Reprinted from Bureau of Mines Information Circular 7462."

^{2/} Chief chemist, Illinois State Geological Survey.

weakly coking coals; low-ash electrode carbon from coal; new developments in the production of metallurgical coke and the utilization of gas and by-products; briquetting brown coal and bituminous coal without the use of binders; recent engineering developments in gas turbines and steam generators, with special reference to use of coal; German coal-fired heating and cooking stoves; and the commercial extraction of coal with solvents for the production of synthetic liquid fuels or chemical products.

Numerous reports on the activities of German synthetic liquid-fuels plants and the plants for the production of synthesis gas from solid fuels are available in the same office as the solid fuels reports.

It is planned to publish in this series of Information Circulars the more important reports or combinations of reports prepared by the members of the Solid Fuels Mission. A similar series on synthetic liquid-fuel activities is being prepared.

A. C. FIELDNER, Chief,
Fuels and Explosives Division.

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PART I. METALLURGICAL COKE^{3/}

INTRODUCTION

This section consists primarily of three reports written by Dr. A. Thau and translated from German into English by him. The reports are entitled:

1. Metallurgical Coke from Coal of Upper Silesia.
2. Metallurgical Coke from Noncoking Coals, with Special Reference to the Two-Step Carbonization Process.
3. Experiences and Suggestions as to the Economy of Raw Materials for the Hydrogenation or Low-Temperature Carbonization of True Coal, As Well As for the Metallurgical Working-up of Small-grained Iron Ores.

^{3/} Prepared from Joint Intelligence Objectives Agency report "Metallurgical Coke, Dr. Ing. A. Thau, Didier Werke A. G., Berlin," 1945. (FIAT Final Report No. 408.)

These reports are reproduced in full as appendices A, B, and C, respectively, at the end of this section.

Dr. Ing. A. Thau, Didier Werke, A. G., Westfälische Strasse 90, Berlin-Wilmersdorf, is a competent carbonization engineer and probably knows more about European developments in the coking industry than any other person.

At the time of the investigators' visit, Dr. Thau had completed the second paper mentioned above and was in the midst of translating it into English. Later, all three of the articles were obtained in English from Dr. Thau. These articles are reproduced in the appendices without change from Dr. Thau's language. Editing would assist in ease of reading and clarity of expression. However, editorial changes were not made in the appendices, as in making such corrections it is difficult to avoid making occasional changes in meaning also.

1. METALLURGICAL COKE FROM COAL OF UPPER SILESIA (ABSTRACT)

Coals of Upper Silesia are geologically young, having a volatile-matter content well above 30 percent and a correspondingly high oxygen content. Narrower coke ovens and a shorter coking time were tried first. Then, compression of the coal charge by means of stamping machines was tried and became almost universal practice in Silesian coke plants. While blending with coals of lower volatile content produces satisfactory metallurgical coke, Silesia's geographical location prohibits any extended use of low-volatile coals because of their high delivered cost. When semi-coke obtained by low-temperature carbonization is blended with Silesian coal and the charge is stamped, a very satisfactory metallurgical coke is obtained. It is essential that care be taken in the preparation of the semi-coke to obtain a product that will blend satisfactorily with the coal used. Dr. Thau recommends the continuously operated vertical chamber oven for low-temperature carbonization.

2. METALLURGICAL COKE FROM NONCOKING COALS, WITH SPECIAL REFERENCE TO THE TWO-STEP CARBONIZATION PROCESS (ABSTRACT)

A summary of this article, as prepared by Dr. Thau, is given at the end of Appendix B.

3. EXPERIENCE AND SUGGESTIONS AS TO THE ECONOMY OF RAW MATERIALS FOR THE HYDROGENATION OR LOW-TEMPERATURE CARBONIZATION OF TRUE COAL AS WELL AS FOR THE METALLURGICAL WORKING-UP OF SMALL-GRAINED IRON ORES (ABSTRACT)

A summary of this article, as prepared by Dr. Thau, is given at the end of Appendix C.

APPENDIX A

METALLURGICAL COKE FROM COAL OF UPPER SILESIA

Introduction

The coals of Upper Silesia consist of geologically young deposits with a content of volatile matter well above 30 percent. Coals having progressed farther in their coalification with a correspondingly lower content of volatile matter have been traced in deeper seams which have not yet been opened.

The available coals have the properties common to most geologically young coals, and high contents of oxygen dominate. The content of volatile matter is rather high and consists partly of bitumen having a low boiling point, so that a great portion of the bitumen is being evaporated before the formation of the plastic seams commences, and in consequence there will be no, or very little, coherent coke.

Thus, the first measure taken to improve the quality of the coke was the introduction of silica coke ovens with narrow chambers, and by the application of high temperatures the coking time is considerably shortened owing to the rapidly penetrating heat. A better coke is being formed, though its pieces are correspondingly shorter because of the narrow width of the coking chambers. The coke, however, is practically free from spongy pieces. A further means adopted to assist the formation of a coherent coke is the compressing of the coal charges by means of stamping machines, which have been introduced in almost every coke-oven plant in Upper Silesia.

In order to distribute the bitumen, when it becomes plastic, over larger surfaces formed by the coal particles in the charge of the coke oven, mixing with coals having lower contents of volatile matter has been tried and practiced with good results at many plants. The unfortunate geographical situation of Upper Silesia in relation to other coal deposits prevents the importation of lean coals suitable for this purpose on account of the excessively high freights, and only few coking plants are so placed that they can adopt this method to improve their coke.

When, at the end of the first world war, the low-temperature carbonization of coal was introduced in Germany, the coal to be mixed with the high-volatile coking coal was, in the Sarre, mixed with semicoke, which in these cases serves as a lean coal, the contents of volatile matter of which have been artificially reduced by this process.

This method was copied in several coking plants in Upper Silesia with much success after the proper conditions had been determined and maintained exactly.

Among several reports which have been published about this problem may be mentioned the paper by F. Boennemann (Gluckauf 1926, p. 1551). The coke manufactured at a particular plant in Upper Silesia was made from a coal having 34 percent volatile matter. It was crushed by means of bar disintegrators and carbonized in coke ovens with chambers 550 mm. wide, being heated

to a flue temperature of 1,200° C. The drum test of the coke came out to 40.8 percent above 40 m. In discarding the bar disintegrator in favour of a mill and applying a modern silica-coke oven with a chamber 350 mm. wide, enabling the application of a flue temperature up to 1,375° C., the coke made from compressed charges yielded 61.2 percent above 40 mm. by the drum test. By adding a semicoke still containing 17 to 19 percent of volatile matter, the coke made from a compressed-coal charge in the narrow silica chamber improved to 80.2 percent above 40 mm. by the drum test. The exact conditions of numerous tests carried out in this connection, as well as all values and figures relating to these trials, can be deduced from the published article cited above.

Several years later, H. H. Koppers occupied himself with the same problem in Upper Silesia. (Dr. Ingenieur Dissertation, T. H. Aix la Chapelle.)

The trials carried out by H. H. Koppers were on lines similar to those mentioned above, except that the coke ovens had a medium width of 450 mm. The coke normally made yielded an average 41 to 60 percent above 40 mm. by the drum test. By admixing 15 percent semicoke, the results of the drum test could be improved and equalled 68 to 73 percent above 40 mm. by the drum test. This was satisfactory, because the claims made in Upper Silesia on the properties of metallurgical coke correspond to 65 to 70 percent above 40 mm. by the drum test.

Semicoke as an Admixture to Coking Coal

The admixture of finely powdered semicoke to the coking coal offers the advantage that the material can be produced on the spot without incurring additional freight charges. The many failures in connection with the method of mixing semicoke with coking coal must be attributed to the fact that, in subjecting the coal to low-temperature carbonization, insufficient care has been taken to produce a coke of special quality suited to this purpose. Low-temperature carbonization plants are generally operated to obtain the highest possible yield of primary tar, but the semicoke produced under these circumstances is not suitable for admixture to coking coal. The coal must be treated by applying even heat throughout, and a continuous process keeping the coal in motion all the time is preferred to obtain a semicoke of absolutely even properties, free from zones of different quality. The semi-coke must be active; that is, it must participate in the carbonization of the coking coal, otherwise it will behave like an inert material in the coal charge and will yield a very friable coke.

Personally, I have adapted the continuously operated vertical-chamber oven for the low-temperature carbonization of noncaking or slightly caking coals, giving the chamber a medium width of 200 mm., and the heating of both side walls is effected by means of hot circulating gases, which assure the distribution of an absolutely even temperature over the entire wall surface. Excellent results as to throughput, properties of semicoke, yield, and quality of primary tar have been obtained on a commercial basis. As the oven is constructed of fire-clay bricks, and as comparatively low temperatures are applied, the oven will last a very long time, and many times longer than the ovens constructed of metal for the same purpose.

As a reliable guide for the desired quality of the semicoke used for this purpose, its contents of volatile matter are kept under close control all the time.

APPENDIX B

METALLURGICAL COKE FROM NONCOKING COALS WITH SPECIAL REFERENCE TO THE TWO-STEP CARBONIZATION PROCESS

Although the population of the earth has, since the beginning of the 19th century, grown by almost three times in number, the production of coal has increased 100 times and the manufacture of iron nearly 200 times. Since the production of 1 ton of pig iron requires at least 1 ton of metallurgical coke, it is obvious that the manufacture of coke also has increased in equal proportion. By the ever-increasing proportion of coal subjected to carbonization in relation to the total output of the coal mines, a distinct shortage of good coking coal is acutely felt in all industrial countries, and different means are resorted to in order to counteract this serious deficiency.

Suggestions by L. Weber

Remarkable suggestions aimed at alleviating the shortage of coking coal have been forwarded by L. Weber (1)⁴ to the effect that noncoking fines are mixed in a predetermined proportion with fine-grained iron ore, and, by the addition of a suitable binding medium, the mixture is pressed into briquettes. These are subjected to low-temperature carbonization to harden the structure and to recover the valuable low-temperature tar. The amount of carbon contained in these briquettes or ovoids suffices for the chemical reduction of the ore mixed with the coal. The briquettes are melted either in low-shaft furnaces or in liquid-slag gas producers, recovering at the same time the gas formed. The practical application of these rather bold suggestions, which, as has been amply proved by numerous trials, can, without technical difficulties, be readily materialized, would place the whole manufacture of iron in its present form on a new basis, because the blast furnace as well as the coke-oven plants would disappear entirely to give way to low-temperature carbonization plants and low-shaft furnaces or liquid-slag gas producers.

Such a revolutionizing change in pig-iron manufacture naturally can not be introduced within a short time, and its manyfold effects require careful consideration to guard against disappointments. In spite of this natural reluctance, which will impede the introduction of such radical changes, these remarkable ideas have been mentioned here to show that the want of good coking coals in all industrial countries brings forward suggestions for the manufacture of pig iron without the use of metallurgical coke.

Widening the Range of Coking Coals

The presence of a sufficient proportion of bitumen with agglutinating properties in the coal makes the formation of a hard and lumpy coke a foregone conclusion. The agglutinating bituminous parts of the coal are those which

⁴ Figures in parentheses refer to items in the bibliography at the end of part I.

justify the name of coking coal. As long as caking coals were available in abundance, there was no need to bother about the agglutinating power as the primary property of the coking coals. Since that portion of the coal mined as suitable for coking coal has been for a long time rather small to cope with the demand of the coke-oven plants, the agglutinating properties of the available coals require great attention, and the well-coking coals are being mixed with weakly-coking kinds to stretch their quantity and also to improve the quality of the coke, in many cases, by these means.

Depending upon the quality of the caking coal, the proportion of the coal, with minor agglutinating power that can be added remains in many instances, rather small, and a severe crushing as well as an intense mixing of both kinds of coal is of particular importance to produce a coke of high quality. The details of this treatment will, beyond a hint to the respective literature (2), not be considered here. In this connection, it should be mentioned, however, that admixture of 10 to 15 percent lignite suffices to eliminate the agglutinating properties of the mixture altogether.

Attempts to Manufacture Coherent Coke

As far back as several decades ago, trials were carried out in England to produce a coherent coke from noncoking coals by the addition of a certain amount of hard or soft coal-tar pitch before carbonizing. In technical respects, this process has proved its adaptability, but for economic reasons it was a complete failure, and for this reason the process has, apart from individual trials, never been introduced on a commercial scale. If the pitch is equally distributed over the whole charge of coal, which latter must be reduced by grinding to a very fine state, a coke of evenly dense structure will be formed, as has also been proved by Thau (3), in carrying out trials along the same lines.

Since coal and pitch can, on an industrial scale, be intensely mixed only with difficulty, thorough mixing of coal and oil is imperative for the formation of a hard coke structure, even if the hard or soft pitch is added in the liquid or granulated state. New ways have been adopted in recent times for the same purpose, and the "coal in oil" process was developed, the details of which are described elsewhere (4).

Oil and tar can, particularly in the heated state under which they are converted into a very fluid liquid, be easily mixed with coal, and during carbonization the low-boiling portions are evaporated while the high-boiling residue is carbonized with the coal and causes the coal particles to be cemented to each other.

Several low-temperature carbonization processes have been developed along these lines, but it is not intended to dwell upon their details (5) in this connection.

During the development of the modern low-temperature carbonization processes, the problem of manufacturing a coherent semicoke was again given prominence, after experience had proved that without producing at the same time a hard, dense, and lumpy semicoke these processes would not from an

economical point of view, be able to justify their existence. Since in many instances the hardness of the semicoke, even if manufactured from good caking coal, left much to be desired, in some plants the coal was placed into molds, which were then subjected to low-temperature carbonization. By these means, a molded, evenly shaped semicoke is produced; but these processes (6), mentioned here only incidently and of which several have been developed in England (7), had no lasting success.

Since it is one of the purposes of low-temperature carbonization to work up the cheap noncaking or weakly caking coals for which the conditions of forming a coherent coke are considerably more unfavorable, compared with carbonizing, at high temperatures, special means had to be adopted to assist the formation of coke, such as are already indicated above. In these processes the missing or too low-boiling bituminous constituents are to be supplemented by addition of pitch or tar. F. Fischer and his collaborators (8) investigated several low-temperature carbonization processes developed in France along these lines, mixing the coal before subjecting it to low-temperature carbonization with tar, anthracene oil, pitch, or coal-tar oil. These research workers eventually adopted different ways of applying a binding medium which is being produced in the low-temperature carbonization plants themselves. The low-temperature tar was found to serve this purpose, although, if once applied, it loses its binding property and, if used further for this purpose, the tar must be oxidized by blowing air through it. By these means a coherent hard coke with even structure was formed by low-temperature carbonization, and at the same time 80 to 87 percent of the tar added to the coal was recovered. However these trials have not been made on a commercial scale and, as will be noted from the following, quite different means have been adopted for the purpose.

Fuelite Process

In England, Strafford (9) developed the "Fuelite" process (10) as a result of his efforts to convert noncaking or weakly caking coals into a coherent coke. The first step in this direction was a process to manufacture an easily combustible fuel from fine-grained anthracite or coke mixed with coal tar. The residual pitch of the tar serves as a binder for the coal particles, while the oil constituents are evaporated during carbonization. The fuel to be carbonized is first crushed very finely and subsequently is mixed with a barium salt, barium carbonate being the most suitable. An additional admixture of finely powdered charcoal is of advantage. This fine-grained mass is mixed with a certain proportion of coal tar large enough to bind the coal particles, and after the charge has been thoroughly mixed, it is carbonized at temperatures between 400° and $1,000^{\circ}$ C., depending on the quality of the coke to be produced.

The presence of the barium carbonate in the charge has a catalyzing effect upon carbonization, and it also shortens the coking time. As an example for the mixture to be applied, it is stated that dry steam coal or anthracite, coke breeze, or dry coke dust can be ground either individually or as a mixture. At the same time, a finely powdered mixture is prepared, consisting of two parts barium carbonate and three parts charcoal, and to this is added 3 to

12 percent coal dust. Of this mixture, consisting of coal, charcoal, and barium carbonate, about 5 percent is added to the charge and intimately mixed. The charge now receives an addition of coal tar in such a proportion that the single coal particles are covered by it and will stick together. In this condition, the charge is being carbonized, and the height of the temperature applied depends on the kind of fuel it is intended to produce.

The fuel manufactured in this way is significant for its high calorific value; it burns without forming smoke and is easily ignited by wooden sticks. The fuel can be used in domestic fireplaces and ovens of any kind, and after combustion a fine-grained ash remains as a residue. The tar recovered by this process is claimed to be free from pitch and dust, and it consists of oils only, so that it can be separated into single fractions by plain distillation. The truth of this statement must be doubted, as the formation of pitch and dust in coal tar depends, to a large measure, on the kind of carbonization process and the temperature applied. In any case, there is no reason to suppose that the admixtures to the charge mentioned above would reduce or eliminate the thermic decompositions, which are unavoidable in connection with certain carbonization temperatures.

A process developed later by Strafford and Pick (11) aims at the manufacture of a coherent coke from noncoking coals. The process has been tried at a gasworks in London, and by the name of "Fuelite" it became known in England.

In this process, the well-known means are adapted to mix a highly bituminous coal with noncoking coal in such proportion that the excess of bitumen over and above the coke-forming requirements is taken up by the lean coal, so that the whole mixture will form a coherent coke. Depending on the conditions applied, a foundry coke, a metallurgical, or a domestic coke may be manufactured by this process. These already well-known means have been improved by the inventors in this process, mainly by mixing the particles of coal very intensively. To accomplish this, the very finely crushed caking coal is slaked with water, soap lather, creosote or similar easily emulsified liquids, so that an emulsion is formed which keeps the very finely powdered caking coal in suspension. This is mixed with the finely ground lean coal. The charge thus prepared is compressed into iron boxes, the dimensions of which correspond to those of the retorts into which the boxes are to be transferred to carbonize their contents. The liquid added to the coal serves a double purpose - first of all, to facilitate the intense mixture of both kinds of coal, and, second, to exert a binding effect upon the particles of coal exposed to the heat.

As an example for the proportions to be mixed, it is stated that 10 to 20 percent of caking coal is mixed with 80 to 90 percent of lean coal. The proportionate amounts depend on the agglutinating power of the caking coal, and the lean coal may have to be ground and mixed in equal proportions with the caking coal in some extreme cases. The caking coal is crushed to a grain size below 1 mm., whereas the grain size of the lean coal may reach 2 mm. The finely ground caking coal is being mixed with enough of one of the before-mentioned liquids to form a suspension; and for the most favorable proportion of liquid to be added, experience has shown that the moisture contained in the

final charging mixture should be about 15 percent. The compressed charges are carbonized at a temperature between 600° and 1,200° C., and, depending on the temperature applied, a lumpy domestic coke or a hard metallurgical coke suitable for consumption in the blast furnaces may be produced.

Based on the present state of carbonization techniques, the process offers the possibility of producing a coherent coke by the means indicated above. Taking into consideration, however, the capital charges, the operating costs, the heavy wear and tear, as well as the comparatively large consumption of power which the crushing, mixing, and compressing plant requires, the result will, in most cases, exclude the practical application of this process for want of economy. In scientific respects, however, the indicated methods are very interesting, and in exceptional though probably rare cases the adoption of the process may be justified, but such an instance is not yet known.

Lignite Coke

The overproduction of the small-grained semicoke, the so-called "grudecoke," from lignite induced to trials, which commenced many years ago, to manufacture a hard, lumpy coke, though the high contents of oxygen in the lignite excludes the formation of a coherent structure under the application of heat in a way similar to true coal. Briquetting of the small-grained lignite coke has been applied repeatedly but cannot be considered a satisfactory solution of the problem.

The trials by F. Fischer and his collaborators, mentioned above, induced Staemmler (12) to apply similar means for obtaining a hard, coherent coke from lignite. He used lignite gas-producer tar, which in the crude as well as in the oxidized state he mixed intimately in the proportion 1 : 1 with the lignite, which was then subjected to low-temperature carbonization. Although these trials were carried out on a small scale in the laboratory only and under conditions that assure a much more intense treatment, compared with operations on a commercial scale, it was not possible to manufacture a coherent coke by the means adopted.

All efforts to produce a coherent coke from direct lignite failed and the only solution of this problem was the briquetting of the lignite before carbonizing. In this direction, R. Delkeskamp (13) had already shown the way many years ago by developing his so-called colloidal briquetting process, which treated raw lignite and thus saved the expense of drying. The briquettes produced were subjected to distillation at an exceptionally low temperature in special ovens (14) to harden them and to recover the tar at the same time. This process, which, however, has nothing whatever to do with colloids, was never introduced on a commercial scale and can only claim to be of historical interest, for which reason it is not further taken into account in this connection.

The normal lignite briquette made in the sausage press disintegrates after carbonization, and only by the aid of the ring roller press, applying a pressure of over 2.5 tons/cm², briquettes having a fine-grained structure are manufactured and are so hard that also in the form of coke they retain their shape completely, except for a certain all-around shrinkage. The

particulars about this briquetting process (15) are so well known that they need not be repeated here, and, as to the operation of a plant manufacturing briquette coke from lignite, attention is drawn to a report published by Landwehr (16), which contains all information of importance.

Already the fact that the coherence of the lignite coke depends on the mechanical pressure applied in briquetting, causing the particles to form a close structure without being cemented together by the effects of thermic decomposition of the hydrocarbons, as in the carbonization of true coal, tends to show that, in regard to structure as well as to its formation, coke made from lignite and from true coal, respectively, are two entirely different fuels. Hock and Schrader (17) investigated lignite cokes in comparison to metallurgical coke, and since the chemical composition depends solely upon the quality of the coal from which the coke has been made, only the abrasion test is taken into account to indicate the difference, as shown in the following table.

Drum abrasion test of lignite coke and metallurgical coke

Size above 15 mm.

Kind of coke from -	Lowest value, percent	Highest value, percent	Average of ten tests, each, percent
Lignite	81	96	88
True coal ..	99.0	99.3	99.2

Size above 25 mm.

Lignite	39	85	61
True coal ..	99.0	99.3	99.2

In judging the values given in the above table, it must be taken into account that the abrasion of the metallurgical coke determined to be more than 90 percent cannot be taken as an absolute but as a comparative value. Hard pieces of uniform size were picked out and, exactly like the lignite coke, were subjected to the drum test for 1 minute only in each case, sufficient to indicate the pronounced difference of both kinds of coke as regards their abrasive properties. These figures of comparison readily indicate that the lignite coke manufactured from briquettes made by the ring-roller press can never meet the physical properties demanded from a good metallurgical coke. Taking into account the present state of technics, the problem to produce such a coke from lignite can be solved only by means of the two-step process, which is fully described farther on in this treatise.

Carbonization and Low-Temperature Carbonization Under Pressure

The favorable effect of tight packing of the coal particles in the oven charge in connection with the formation of coke is well known. This is indicated, also, by the carbonization of compressed-coal charges in coke ovens, which was introduced many decades ago. Beyond this well-known method, a favorable influence upon the formation of the coke has been observed if heavy weights are placed on top of the charges in the coke oven. Apart from

numerous trials, this process has never been introduced on a commercial scale because of the technical difficulties connected with its operation.

Blayden, Noble, and Riley (18) reported about further trials carried out recently with the object of determining how far a certain pressure to which the coal charge is subjected during carbonization will affect the hardness of the coke produced. It was observed that pressure up to about 2.9 kg/cm^2 (41 pounds per square inch) was sufficient to produce a hard, coherent coke from weakly coking coal. In continuing these trials and increasing the pressure of the load up to 36 kg/cm^2 (512 pounds per square inch), further improvement of the quality of the coke could not be observed. G. E. Foxwell and E. C. Evans (18) referred to these trials and stated that although there is no object of greater importance in connection with the manufacture of coke, the application of mechanical pressure on the coal charge during carbonization in the coke ovens requires technical means that will impair the economy of the coke manufacture so much that they must be ruled out. A great service would be rendered to the iron and coal industry by the introduction of such a process, which would satisfy in technical as well as in economical respects, as good coking coals are becoming very scarce. As a way out, it was recommended that the coal be subjected to briquetting and that the briquettes, with their dense structure, be carbonized. Good experience has been gained by such trials.

The effect of pressure upon the formation of the coke structure rests with the fact that the coal, while heated under the exclusion of air, temporarily assumes a plastic state, and this at a time just before evaporation of the lower boiling parts of the bitumen commences. This temporary plastic state of the coal can be utilized to produce briquettes by the application of pressure, and continue to subject the briquettes formed to low-temperature carbonization without interrupting the heat treatment of the coal.

Such a process has been developed by Tormin (19) in the shape of his oven with vertical disks for the low-temperature carbonization of coal, which in one step yields a briquetted coke. The process has not been introduced on a commercial scale, however, and the particulars about this construction must be read in the technical literature cited.

The Coppée Co. in Belgium (20) treats the coal by low-temperature carbonization in a continuously operating belt-conveyor oven, and the fuel is discharged at a temperature at which the coal has assumed a plastic state. The discharged fuel drops into briquetting presses, in which briquettes are being formed from coal still in the plastic state. The fuel thus produced can hardly serve as a semicoke, as it is discharged just at the moment when low-temperature carbonization is to commence.

Differing from these processes, two more have become known in which, though they also apply pressure during carbonization, this pressure is applied not to the coal charge but to the coal gas liberated by the heat in the retort. Successful trials with such processes have been carried out independently on a small scale by F. Fischer and R. von Walther together with their respective collaborators. The trials, which W. Fuchs (21) reported, aimed at the manufacture of a coherent coke and were successful, but, considering the technical difficulties connected with a carbonization process

of this kind, it is obvious that their introduction on a commercial scale has not yet been realized.

Single-Step Process Producing Coke from Briquetted Coal

As briquettes made from noncoking coals by the addition of a binding medium in roller presses are not sufficiently hard and therefore can not be used as blast-furnace fuel nor as a smokeless domestic fuel, the briquettes frequently are subjected to a thermic treatment with the object of hardening them, and by these means they are transformed into coke. Their abrasive properties are improved, and by expelling the tar the volatile-matter contents also are reduced. Thermic hardening of the briquettes can be effected in chambers heated from the outside as well as in shaft ovens through which hot gases are circulated as heat carriers. Both kinds of heat transfer are frequently applied to the briquette charges at the same time.

Pieters (22) developed a process by which coal or briquettes from non-coking coals are subjected to low-temperature carbonization in vertical, ceramic, chamber ovens heated from the outside, and in the lower zones high-temperature carbonization is being applied. If treating noncaking coals, Pieters (23) mixes the small-grained coke with pitch as a binder, and the ovoids made from this he subjects to thermic hardening in the same ovens. This last-mentioned process comes under the heading of the two-step processes, which are described in detail farther on in this treatise. Plants operating the Pieters process have been constructed in Belgium and in France.

In England, the Midland Coal Products, Ltd. (24), introduced a low-temperature carbonization process, applying a shaft oven in which ovoids made from noncoking coal with pitch as a binder are transferred into semicoke by means of hot gases as a direct heat carrier and produced by partial combustion set up in the charge of the shaft.

Briquetting and Carbonizing Processes by L. Weber

A quite important contribution to solving the problem of converting non-coking coals into shaped hard coke was made by L. Weber (25), who, first of all, dealt with the briquetting of semicoke and, instead of the rather expensive coal-tar pitch which, in addition, develops a lot of smoke during combustion, he applied sulfide lye as a binder. Instead of the small-grained semicoke, Weber continued his work by briquetting true coal in the same way and subjecting the ovoids to low-temperature carbonization, producing by these means, depending on the temperature applied and the length of time which the ovoids were allowed to remain inside of the oven, a shaped coke suitable for metallurgical purposes on the assumption that the chemical properties of the basic coal comply with these requirements. The process by Weber has been described in the technical literature so frequently (for instance, by M. Naphtali (26)) that the details need not be repeated here. If, however, lignite is to be converted by these means, a two-stage treatment must be applied by which the lignite is being subjected to low-temperature carbonization in the first stage and the semicoke produced thereby is subsequently briquetted. In a second stage of the process, the ovoids are subjected to thermic hardening.

Weber has worked continually on the further development of his process and, based upon trials carried out successfully, it has been proved that the working up of small-grained or soft iron ores by mixing them with the coal in a certain proportion before briquetting and carbonizing them afterwards, such a process must be considered as great progress in the direction of manufacturing pig iron, all the more because the pig iron, as already mentioned in the beginning (1), can be produced with large volumes of gases for power or synthesis purposes, at the same time discarding blast furnace plants and coke ovens altogether. This development will, however, not be considered further in this connection.

A basic condition for the process by Weber is the already mentioned application of concentrated sulfide lye as a binding medium and the addition of slaked clay in a small measure, which serves to strengthen the ash skeleton of the ovoids. The principle process of briquetting has been protected in Germany by the DRP. Nos. 409 550, 424 631, 425 541, and 443 679; it should be mentioned that in patent No. 425 541 the manufacture of very hard briquettes for metallurgical and, particularly, blast-furnace purposes is suggested, in such a way that to the coal to be briquetted parts of the blast-furnace burden are added, which have a hardening effect upon the briquettes. Patent No. 476 319, by L. Weber, protects a process for the manufacture of briquettes particularly from fuels and also from soft ores, blast-furnace dust, etc., by applying a mixture of sulfide lye and inorganic substances as binding medium. Also, in this instance the process proceeds in two steps.

In a patent application, No. W 100 063, of December 9, 1936, Weber applied for a patent of a process for the production of shaped semicoke and low temperature tar from fine-grained fuels, particularly noncoking coal, after thermic pretreatment and briquetting by means of a binder, preheating the coal in the presence of neutral gases, and adding a binder to increase the agglutinating power and using continuously operated low-temperature carbonization ovens. The briquettes are to be carbonized at a rapidly rising temperature.

As an example of the operation of this process, an Upper Silesian gas coal is mentioned, the contents of volatile matter which exceed 35 percent and which, after crushing to a grain size of 2 mm., is heated in a current of oxygen-free gas at a temperature between 250° and 300° C. By this treatment, the larger portion of the oxygen contained in the coal is liberated in the form of carbon dioxide and constitutional water. The coal thus preheated is mixed with coal-tar pitch having a softening point exceeding 65° C., and with this binding medium the coal is being briquetted. The briquettes are subsequently subjected to low-temperature carbonization at rapidly rising heat under the normal temperature, so that besides the valuable low-temperature tar, a hard and coherent semicoke is being produced.

Though the sulfide lye mentioned above together with the small addition of a slaked-clay suspension represent a splendid and, above all, smokeless-burning binder, Weber has, as will be gathered from the last-mentioned patent application, applied a series of different binders to meet various conditions; and, apart from sulfide lye, tar and oil as well as their distillation

residuals and coal partly hydrogenated have been used to broaden the range of application of these processes in an economical direction also.

As already mentioned, the processes by Weber have in the meantime been developed far beyond the limits stated above. The trial results and the experience gained in this connection encourage the conclusion that these processes will in all probability find an important place within the heat economics of the iron industry before long.

Processes for the Manufacture of Shaped Coke in Two Stages

The process for manufacturing shaped coke by subjecting the fuel to a two-stage treatment in succession owe their existence to the efforts to produce a hard and dense, smokeless burning coke of even shape within the range of low-temperature carbonization so as to recover at the same time a valuable low-temperature tar. The first process of this kind, which was already in operation on a large scale in the United States during the first world war, has become known under the name of "Carbocoal" process (27). The details of this plant have been published by Thau (28), and the fuel named "carbocoal" had a good market in the United States. The difficulties experienced in the operation of this plant have been published by Curtis and Chapman (29). These technical difficulties largely impaired the economical results of the plant, so that soon after the conclusion of the first world war the plant was shut down and dismantled. Since this was the first process operated on two stages and introduced on a large commercial scale producing solely a good hard coke of even shape as well as low-temperature tar oils and gasoline, the details, as far as they serve as a starting basis for modern developments in this connection, will be outlined briefly.

Carbocoal Process

In the first stage of the Carbocoal process, the crushed coal is, regardless of its other physical properties, subjected to low-temperature carbonization, but a high yield of tar is desirable to affect the economical results favorably. The coal should have no or little agglutinating power. In the plant mentioned, low-temperature carbonization was effected in horizontal retorts united in pairs, side by side, and they were heated from the outside. By means of propeller blades fixed to two parallel horizontal shafts mechanically driven, the coal was continually conveyed through the retorts.

The semicoke discharged was crushed and mixed with pitch recovered from the distillation of the low-temperature tar. The mixture was transformed by roller presses into ovoids and subsequently was carbonized in inclined chamber ovens at high temperatures. The inclined chamber ovens of usual construction were divided into an upper and a lower chamber by an intermediate brick sole, and both chambers of each oven were heated by common flues and had common doors on both ends.

The process was protected in Germany by patents 382 386 and 392 796, which were applied for in 1920. In the same year the two applications S.52 294 and I.20 407 were handed in at the German Patent Office. In the patent specifications the following claims are made: Process for manufacturing

smokeless-burning solid briquettes in heating the coal first of all to about 440° to 650° C. to reduce its contents of volatile matter to 17 to 11 percent. The semicoke is then mixed with pitch and pressed into briquettes, which are subsequently subjected to a temperature up to about 1,010° C. In a further claim, also, an admixture of lean coal or enriching material is mentioned to influence the formation of the structure of the briquettes.

Among the processes that have been developed to transfer the coal directly into a smokeless-burning shaped coke, the Carbocoal process, without a doubt, holds a fundamental position, and on a large industrial scale it has proved its technical adaptability apart from the fact that in the construction of several parts of the plant grave mistakes have been made. The truth is that a coke in the form of carbonized briquettes was made from noncoking coal, however, at costs that prevented its use for economical reasons in the metal industry. For this reason, the process was confined to the single plant erected in the United States which, as already mentioned, has been dismantled since without having been copied on a similar scale elsewhere.

Shaped Metallurgical Coke Manufactured in Two Stages

It was a foregone conclusion to adopt processes resting on the same or a similar basis for the manufacture of shaped coke suitable for use in the metal industry. Such a process offers the great advantage that it is quite independent of the physical properties of the coal and also of the agglutinating power. A primary condition is only a content of tar high enough to supply the quantities of tar pitch required for briquetting the semicoke. Apart from the quality of the coal and of the semicoke produced in the first stage, the desired properties of the final shaped coke leaving the second stage can be influenced considerably by the operating methods of the last stage, so that the demands as to specific gravity, porosity, abrasive properties, ignition point, and reactivity can be met easily. As has been proved by practical trials, the shaped coke produced by these means is highly suitable as a blast-furnace fuel, and as the even shape of the pieces reduces the loss by abrasion in the blast-furnace burden considerably, its application for this purpose is quite justified and also recommendable for economic reasons, even if its price should be slightly higher than that of ordinary metallurgical coke.

Swietoslawski and Chorazy (30) reported on trials made to produce a coherent semicoke or a metallurgical coke from noncoking coal. The coal was mixed with 4 to 10 percent of pitch and made into briquettes at a pressure of 250 kg/cm² and subsequently carbonized. For the quality of the shaped coke thus produced, certain physical conditions must be taken into consideration, particularly in reference to the manufacture and heating of briquettes, as this is of particular importance. Apart from this, the quality depends also upon the composition of the basic coal, though this influence is largely eliminated by the semicoke in the intermediate stage. Coke from very weakly coking coals excels in hardness. As to the ignition point and reactivity, the shaped coke with its very small pores is much superior to the metallurgical coke from Ruhr coal. These trials evidently have been carried out on a small scale just to prove the technical adaptability of the process without having introduced it on a commercial scale.

Delkeskamp Process

The process developed by Delkeskamp under the name of colloidal briquetting, with subsequent carbonization at a very low temperature for minor fuels, has been protected by German patents under the Nos. 577 766 and 674 649 and has been mentioned before in this treatise. In this connection, the processes developed after the death of Dr. Delkeskamp by his collaborators G. Merkel, W. Groth, P. Jung, and others should just be mentioned as far as they coincide with the aims considered here.

Since the original carbonization oven caused a lot of breakage of the briquettes, a new type was constructed with vertical metal retorts spirally twisted to retard the passage of the small-grained material into which the briquettes are to be embedded while passing through them. These ovens, the details of which need not be described here, were protected by German patents 626 590, 629 599, and 630 592 covering 23 patent claims altogether.

A further process was protected by the German patent 717 324 for carbonizing minor fuels such as lignite, peat, etc., in two stages. The fuels were first subjected to low-temperature carbonization, briquetted after the addition of a binder, and subsequently again carbonized by applying a temperature not exceeding 600° C., so that a reactive semicoke was formed. The semicoke from the first stage was crushed down to about 8 mm., mixed with 20 to 30 percent small-grained caking coal, if necessary, water was added, and subsequently the mixture was briquetted and the briquettes were carbonized as mentioned to produce a fuel suitable as a substitute for charcoal. If a gas coal is added as a coking medium the proportion to be added is somewhat smaller than stated above.

The temperatures mentioned in the patent claims and as applied in the second stage of the process show that this process aims at the manufacture of a semicoke suitable for all purposes for which this fuel is to serve, particularly, also, for automotive-gas producers. The manufacture of a metallurgical coke by the means indicated does not seem to have been contemplated within the range of this process.

Two-Stage Process for Manufacturing Shaped Metallurgical Coke

In the following process developed by the Didier-Werke, the treatment of the fuel is almost the same as by the processes just mentioned, only the final aim rests with the manufacture of a coherent hard metallurgical coke. On the whole, the process represents the uniting of a series of known and proven methods, so that the technical operation does not offer any new problems but simply a suitable co-ordination and a corresponding co-operation of individual stages within the process.

The particular advantages of the process as compared with the operation of coke ovens rest first of all with the selection of the basic coal, as the process is altogether independent of its agglutinating power or other properties. Economically, however, the coal must have so high a yield of tar as to cover the requirement of the pitch as a binder for the briquetting plant

working up the semicoke. As a second advantage, the quality of the coke is to be mentioned which is manufactured in the form of even shapes and which, in respect to pressure resistance, abrasive properties, ignition point, porosity, reactivity, and contents of volatile matter, can easily be adapted for all purposes for which it is to be used.

Basis of Process

In a brief outline, the principle of the process can be described as follows: The coal having a grain size of 0 to 50 mm. is subjected to low-temperature carbonization in a suitable oven at a temperature of 550° to 600° C. and freed from its tarry constituents. A construction particularly adapted for this purpose of the continually operated vertical-chamber oven as applied on many gasworks has been developed and successfully introduced on a commercial scale for this process. The semicoke continuously discharged is cooled and crushed and mixed with 5 to 8 percent of coal-tar pitch and 10 to 15 percent of caking coal. This mixture is made into briquettes by applying, in a roller press, a pressure of 100 to 200 atmospheres to produce briquettes of the most suitable shape and of a sufficiently high pressure-resisting quality. These briquettes are carbonized in continuously or intermittently operated vertical-chamber ovens applying a temperature of 900° to 1,000° C. They are thus hardened and are cooled after discharge. The temperature applied in the first and second carbonization stages, respectively, as well as the time taken for the thermic treatment in each of these two stages, which latter may also be termed "speed of throughput," are responsible for the contents of volatile matter in the final coke, and at the same time the height of the ignition point and the reactivity of the coke, as well as other properties, can be influenced by these means.

Technics of the Manufacture of Shaped Metallurgical Coke

The manufacture of shaped metallurgical coke, first from lignite and subsequently from true coal, will be explained briefly, supplemented by sketches, graphs, and tables. It should be mentioned beforehand that the figures stated refer to results obtained from trials carried out on a commercial scale. First of all, a shaped metallurgical coke produced from a Jugoslavian lignite by this process has been used exclusively for burdening a blast furnace, and this trial has proved the superiority of the shaped coke for this purpose in comparison with the metallurgical coke-oven coke.

The manufacture of coke on a commercial scale from geologically young noncaking fuels such as lignites, and its application for metallurgical purposes was in most cases a failure, for two reasons, viz.: (1) By the chemical and physical properties of a large portion of the basic fuel, particularly by the lack of agglutinating power, and (2) for lack of economy of the processes applied.

The development that has taken place in recent times in the operating technics of blast furnaces and coke ovens is responsible for the fact that these problems have been solved successfully and methods have been applied that take care of the economic and other necessities as presented by local conditions in different cases.

Kinds of coke produced at temperatures between 500° and 600° C., in most cases made from inferior fuels, characterized by a complete freedom from tar and by high reactivity, have repeatedly proved the advantage of their application for the chemical reduction of metal ores on a large commercial scale and have also shown good results of the electric blast furnace. The coke used for direct chemical reduction can be applied in the small-grained size just as manufactured without briquetting and subsequent thermic hardening, so that for its manufacture only the first operating stage of the process will be necessary, as no particular demands are made on the physical properties of the coke for this purpose, contrary to those of the coke used in the blast furnace, which, manufactured from geologically young fuels, demands special means, attention, and experience for its production.

The problem in manufacturing a suitable blast-furnace coke from lignite or other geologically young noncaking fuels was suggested from quarters in some foreign country lacking good coking coal, and the Didier-Werke took the hint and solved the problem in a practical and satisfactory manner after numerous trials on a small and large scale. This work has been concluded by a blast-furnace trial run covering several days and giving the absolute proof that the metallurgical coke made from lignite by this process was in every respect a suitable blast-furnace fuel.

Basic Fuels

For the manufacture of metallurgical coke by this two-stage process, nearly all minor fuels, such as brown coal, lignite, xylite, and other non-caking kinds of coal having a correspondingly high content of tar to satisfy the requirements of pitch for the production of the briquetting plant of the process, are readily suitable. As to the physical properties that are desirable in the manufacture of metallurgical coke by this process, a lumpy or rough-grained condition, as well as a certain resistance of the structure to heat treatment, is the only demand to which nearly all fuels readily comply. In addition to the grainy and lumpy fuels the small-grained sizes also serve as basic fuel for this process. Their treatment, however, necessitates certain alterations of the ovens of the first stage to deal with fines instead of grainy or lumpy fuels. As to the chemical properties of the basic fuels, all kinds of coal can be treated if their contents of ash and sulfur do not exceed certain limits, so that even widely varying kinds, such as are compiled as outstanding examples in table 1, will come within this range.

As to table 1, which includes the most important lignite deposits of Europe compiled as to their properties, it should be emphasized that the figures are based upon tests of characteristic samples that represent the average of the fuels mined without taking into consideration fuels of exceptional composition. From the values in table 1 can be deduced that the German lignite has the highest contents of water, and this must be taken into account when judging the yield of tar from these fuels. The contents of water diminish at the same rate as the geological age of the lignites increases, while at the same time the contents of carbon rises as a characterizing effect of the gradually progressing coalification. The three lignite deposits of Bohemia have the highest contents of tar, though this must, to a certain extent, be attributed to their low contents of water. Thus, in order to create a better basis of comparison, the decisive values of table 1 are also inserted in relation to dry fuel.

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TABLE 1. - Properties of lignites of different origin, percent

	Rhenish	Central Germany	East Elbian	Falkenauer	Komotauer	Dux- Brüx	Jugo- slavia	Hungaria	Bulgaria	Greece	Rumania
<u>Proximate analysis:</u>											
Water	50.0	50.0	50.0	28.1	35.2	27.8	48.0	18.6	41.4	47.1	11.0
Ash	3.2	4.4	2.7	10.1	7.6	3.1	3.5	11.7	2.1	2.6	6.9
Crucible coke	24.8	24.7	23.5	37.1	34.4	35.3	23.0	44.8	22.1	19.1	47.5
Volatile matter	25.2	25.3	26.5	34.5	30.4	36.9	29.0	36.6	36.5	33.8	41.5
Pure carbon	46.8	45.6	47.3	61.8	57.2	69.1	48.5	69.7	54.5	50.3	82.1
<u>Low-temperature carboni- zation test:</u>											
Water	50.0	50.0	50.0	28.1	35.2	27.8	48.0	18.6	41.3	47.1	11.0
Chemically formed water.	5.3	4.7	6.3	4.7	5.1	5.8	6.8	7.9	7.0	9.0	6.5
Low-temperature tar	4.2	5.3	4.3	13.3	11.6	15.0	6.7	3.7	10.7	9.0	9.5
Semi-coke	29.9	29.4	29.6	45.9	41.2	43.2	29.8	53.1	32.9	25.6	55.5
<u>Based upon dry coal:</u>											
Chemically formed water.	10.6	9.4	12.6	6.5	7.8	8.0	13.0	9.7	11.9	17.0	7.3
Low-temperature tar	8.4	10.6	8.6	18.5	17.9	20.7	12.8	4.5	18.2	17.0	10.6
Semi-coke	59.8	58.8	59.2	63.8	63.5	59.8	57.3	65.2	56.1	48.4	62.3
Ash	6.4	8.8	5.4	14.0	11.7	4.3	6.7	14.3	3.5	4.3	7.7
<u>Based upon dry coke:</u>											
Ash	12.9	17.8	11.5	27.2	22.1	8.8	15.2	26.2	9.7	13.8	14.6

There are, of course, certain qualities that make it desirable to discriminate between the available lignites to be used for manufacturing metallurgical coke, and the properties pointed out in the following require attention.

Size of Grain and Hardness

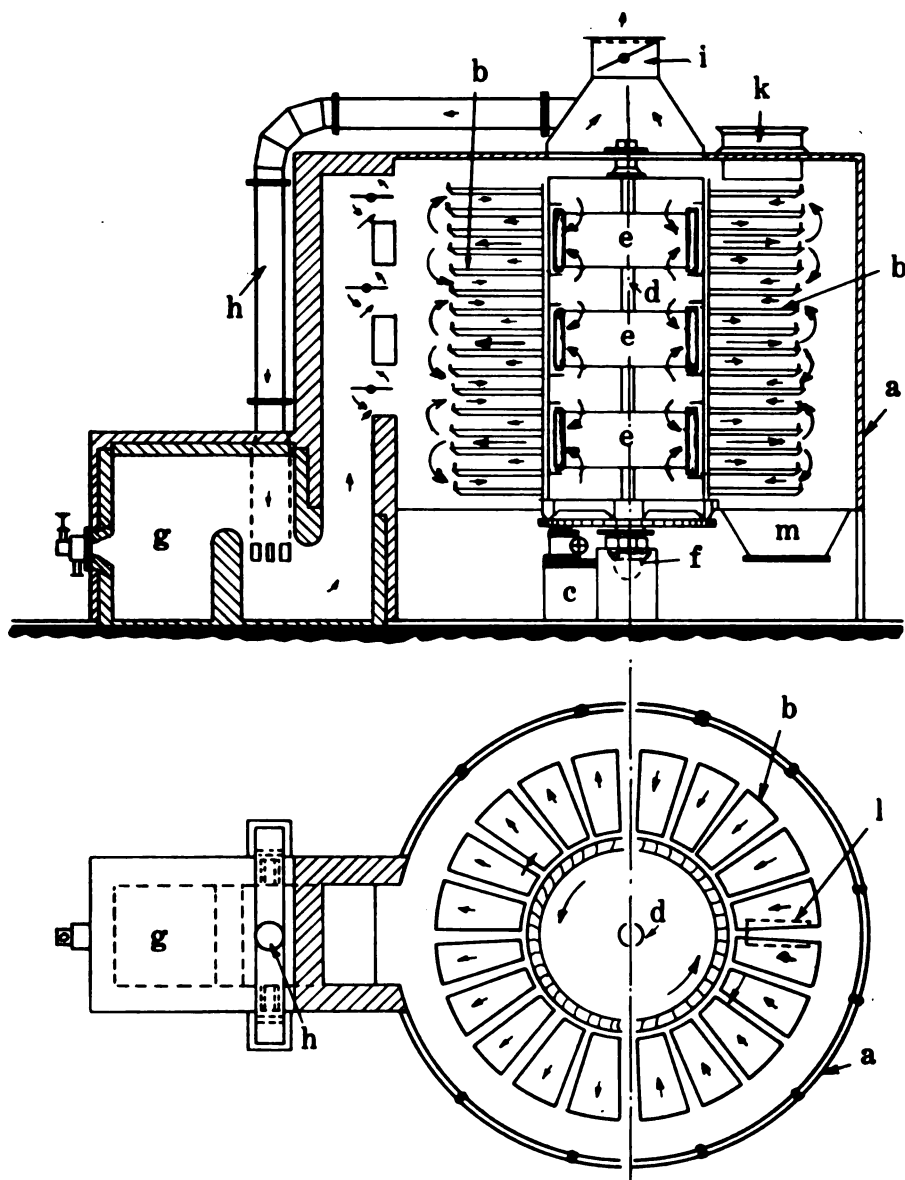
The demands made on the basic fuel, as regards size of grain and hardness, can easily be met, especially as almost every fuel can readily be utilized after having been crushed to 0 to 60 mm. Separation of the dust and of the portion 0 to 3 or 0 to 5 mm. is recommended for reasons that will be referred to in connection with the contents of ash farther on. The small grain additionally created by passing the charge through the oven chamber by abrasion in a size of 0 to 5 mm. is so small that it need not be considered.

Contents of Water

In itself, the content of water in the fuel is of no importance in connection with transforming the fuel into metallurgical coke. Fuels containing 15 to 25 or even 30 percent water, which are common, can be carbonized readily. If the content of water exceeds 30 percent pretreatment in a drier is recommended, by which moisture is reduced to 15 to 20 percent. As carbonizing ovens are the most unfavorable apparatus imaginable for drying fuels, and because their throughput is reduced out of all proportion by a high content of water in the charge, in keeping with the latest developments of carbonizing technics, the fuel is dried in separate apparatus, which is interposed before the carbonizing units of the plant. By these means, the throughput of the ovens is increased considerably. Depending on the physical behavior of the fuel, all drying processes available can be applied for this purpose, particularly the driers operated by combustion gases as well as the disk and tubular drum driers, both operated by indirect steam. Instead of the drying process by Fleissner, which was developed to prevent the disintegrating effect which drying frequently exerts upon such fuels, thus offering the advantage of preserving and solidifying the lumps, the turbine drier constructed by Buttner-Werke can, after extensive experience, be used for this purpose all the more, as many such units have been introduced on a commercial scale.

Turbine Drier

This drier, shown in horizontal and vertical section in figure 1, has proved its value on a large commercial scale in many plants, and it fully meets all technical and physical demands made on it. As shown in figure 1, a stationary cylindrical housing, a, well-insulated against heat loss by radiation, occupies a framework, b, consisting of ring disks and is slowly revolved by means of the electric motor c. In the center of the framework, b, a vertical shaft, d, is arranged, to which are fixed a number of circulating fans, e, the blades of which are shaped like turbine blades. There is one fanwheel for every stage of the dryer. The shape of these fans is responsible for the name "turbine" drier, by which the apparatus is known. The fan shaft, d, has an independent drive from the gear, f, and is revolved in the opposite direction from the framework, b. If hot waste gases from a near-by source are not available, the combustion gases serving as heat carriers are produced by the



- | | |
|-------------------------------|--------------------------|
| a. Housing | g. Furnace |
| b. Revolving frame work | h. Return of waste gases |
| c. Drive of frame work | i. Waste gas outlet |
| d. Turbine fan shaft | k. Coal feed |
| e. Turbine fans | l. Passage |
| f. Drive of turbine fan shaft | m. Coal discharge |

Figure 1. - Turbine drier.

combustion of powdered fuel or of gas in the furnace, g; and by recirculating and mixing with cooler gases, the proper temperature of the drying gas is adjusted. For this purpose the return pipe, h, is arranged between the furnace, g, and the top of the drier; and in the upper outlet, i, of the latter, a butterfly valve is provided, which facilitates the adjustment of the returned gas volumes. The way of the gases from the furnace through the drier and in between the single-ring disks, guided by the effect of the turbine fans, e, is indicated by arrows in figure 1. The waste gases loaded with vaporous moisture escape through the upper outlet, i, and are conducted through a dust separator before being released into the open air.

The fuel is introduced through the opening k in the top of the housing a at a uniform rate so as to allow for its even distribution on the upper ring disk. Each ring is in one place provided with an open slot, l, and over this a stationary elastic scraper is fixed, which forces the fuel, after having made one revolution on the top with the ring disk, to drop onto the next lower ring disk, on which it is also distributed in an even layer. This operation, commencing on the upper ring disk, is repeated downwards in the drier from disk to disk, until at last the fuel is discharged through the outlet m in the bottom of the drier housing, from which it drops onto a conveying belt, leading to a screening plant. At the same time air is drawn in through the outlet m by means of turbine fans, which causes the dried fuel to cool somewhat. The fuel is treated very gently while passing through the drier, and little dust is formed during drying. The number of revolutions of the framework carrying the ring disks, corresponding at the same time to the turning over of the fuel to which it is subjected while passing through the drier can be very closely adjusted to the original contents of water of the fuel and to the desired moisture contents of the dried fuel. The fuel passes in a vertical direction to the forward and backward gas-current zones of each single turbine fan c, so that the drying gases are conducted in a changing direction over the fuel and exert their drying action.

A particular characteristic of the turbine drier is the very gentle treatment of the fuel in mechanical respects during the drying process as well as absolute control of the heat introduced, and ample means to regulate the temperatures are provided. Under these circumstances, an even drying down to the predetermined contents of water in the dried fuel is effected while the size of the lumps is hardly reduced by the gradually progressing drying. In economical respects, a low consumption of power as well as small losses of waste gases and dust should be mentioned as particular advantages of this drying process. The heat consumption of the drier is covered by the combustion of the fines screened out after crushing the raw fuel, as its removal from the fuel to be carbonized is, as already mentioned, desirable on account of its high contents of ash.

Contents of Ash

Lignites in the raw or predried condition still contain 15 to 20 percent water and, besides this, a certain amount of ash, the presence of which is of particular importance for the manufacture of metallurgical coke. As the yield of coke of these fuels does not, as can be deduced from the table 1, exceed 35 to 45 percent, the contents of ash are increased by carbonization 2.2 to 2.8

times. In carbonizing a fuel containing 5 percent ash, the contents of ash are thus increased by carbonization to 11 to 14 percent. The contents of ash in the coke made from the fuels mentioned are stated in the lowest line of the table 1.

As the ash contained in the coke has a considerable effect upon the operation of the blast furnace, in this connection the principle must be observed, also, to use if possible only fuels with low contents of ash for carbonization. The content of ash in the coke should as a rule not exceed 8 to 9 percent, but the maximum of ash permissible depends on its chemical composition. This point will be dwelled upon briefly hereinafter and as an example of a coke that justifies the presence of higher contents of ash the following table gives the chemical composition of a lignite ash.

Chemical composition of ash in lignite

Constituent	Chemical formula	Percent
Silicon	SiO ₂	10.88
Alumina	Al ₂ O ₃	4.51
Iron	Fe ₂ O ₃	35.68
Magnesia	MgO	1.25
Lime	CaO	42.55
Alkalies	-	5.13

Effect of Composition of the Ash

In most cases the ashes of geologically young fuels differ considerably from those contained in normal blast-furnace coke, to which fact M. Paschke (23) has drawn attention. For comparison of the differences of the composition of ashes, the results of tests made have been compiled in the table 2, referring to characteristic kinds of lignite and of normal metallurgical coke. To judge these values, it should be mentioned that the sulfur contained in the coal, depending on the conditions of the mode of determination, remains the more in the ash the more lime the ash contains.

The effect of the lime in the ash of the coke upon the lime requirements of the blast-furnace burden is shown in figure 2, which was plotted by C. Schwarz based upon the investigations of Mathesius and Endell (24), in which the vertical ordinates represent the contents of alumina, whereas the horizontal abscissas stand for the contents of silicon in the slag. The range of the usual blast-furnace slags when charcoal is used as a fuel, as well as metallurgical coke for producing pig iron for foundry or steelworks purposes, are, as to their properties, framed in on the graph, while the ranges concerning Portland or alumina cement are elevated by section lines. In order to judge easily the liquidity of the slags, the lines representing the viscosity at a temperature of 1,400° C., according to Endell, are put in. At higher temperatures they are to be moved further to the right. Slags which, according to their composition, remain outside the range indicating, in the graph, the favorable kinds, require an addition of lime to the burden if they do come in on the right and above the framed field. In its place, an addition of sand to the burden is required if they appear on the left and below this space, and correspondingly more ore of a basic or more acid nature, respectively,

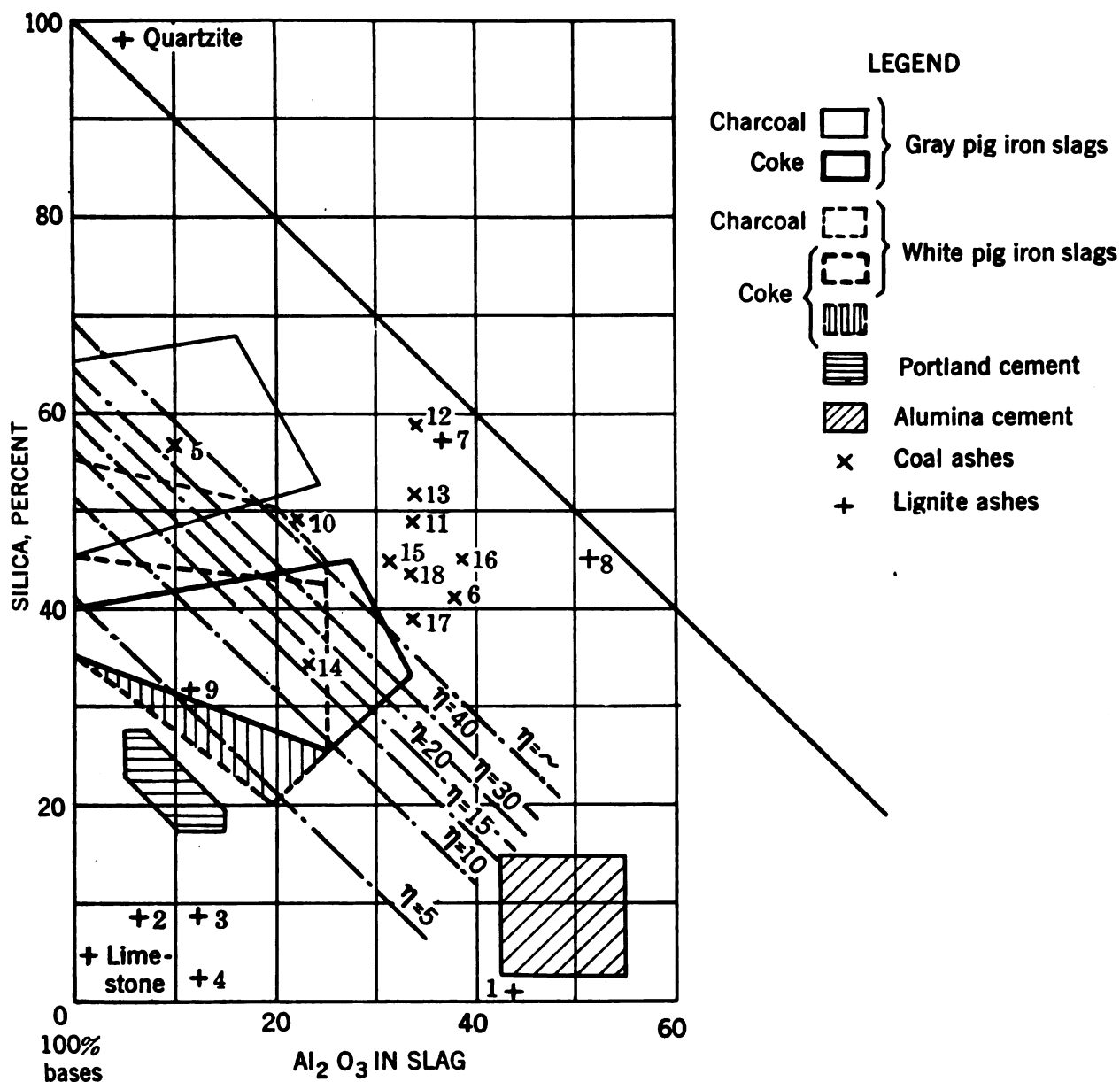


Figure 2. - Composition of coke ash and its effect upon blast furnace operation. According to Endell, at 1,400° C., $\eta = \frac{1.9}{K_Z - 0.45}$ (viscosity, poises), $K_Z = \frac{100 - (\text{SiO}_2 \text{ Al}_2\text{O}_3)}{\text{SiO}_2 \text{ Al}_2\text{O}_3}$

must be added to the burden. To the points in the graph indicating limestone and quartzite, numbers are attached which indicate how many kilos of limestone or quartzite in relation to 1 kilo of slag produced from it must be added. The further figures in connection with these points on the graph correspond to the numbers of the ashes compiled in table 2. From these values can be deduced that the lignite ashes appearing in table 2 and numbering 2, 3, and 4 correspond, in respect to their chemical composition, to a ready blast-furnace slag. The ash appearing under No. 12 comes within the range of the aluminous cement and would be particularly suitable for melting such iron ores as have too low a content of alumina. The remaining lignite ashes appearing in table 2, in respect to their chemical composition, approach very near that of normal coke and require, with the exception of the ash No. 14, all additions of limestone or basic ores. In comparing the contents of ash, it must be taken into account that in smelting acid ores and using lignite coke with a basic ash, a higher content of ash, compared with the normal coke, is by no means a disadvantage. In smelting, in the presence of acid ashes from normal coke, about 1.5 to 2 times the amount of slag in relation to the contents of ash will be produced after neutralizing with lime. Given by weight, it corresponds 1.75 times to the addition of limestone if not, instead of this, basic ores of a corresponding composition are smelted. On the contrary, owing to the presence of basic lignite ashes, practically an equal weight of lime or 1.5 to 1.6 times the amount of limestone can be saved by smelting basic ores.

TABLE 2. - Chemical composition of ashes of different kinds of lignites and of cokes from true coal, percent

Origin	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaC	MgO	K ₂ O+Na ₂ O	S	P ₂ O ₅
<u>Rhenish lignites:</u>								
1. Fortune mine	1.75	40.6	10.58	35.89	10.0	1.18	-	-
2. Average	6	5	15	50	4	-	8	-
<u>Central Germany:</u>								
3. Werschen-Weissenfels district	4.97	5.8	19.05	37.1	1.99	0.83	8.3	-
4. Hofmann I mine, Helmstedt	2.20	9.04	16.30	45.00	5.30	-	5.3	0.98
5. "Alwiner Verein", Halle.	51.05	9.22	5.33	26.57	1.72	1.81	1.7	-
<u>Cassel:</u>								
6. Gewerkschaft Friedr. Rungen	34.09	32.3	9.04	14.26	1.84	0.57	3.1	0.14
<u>Bohemia:</u>								
7. Bohemian lignite	51.2	33.91	9.15	2.64	0.48	1.01	1.0	0.04
<u>Silesia:</u>								
8. Lignite from Muskau	43.2	48.2	7.07	0.52	0.58	0.28	0.2	-
<u>Upper Bavaria:</u>								
9. Pitch coal, Peissenberg	21.37	9.11	22.62	29.21	5.18	-	4.2	1.59
<u>Jugoslavia:</u>								
10. Shaped coke from lignite Kreka mine ...	33.74	15.20	19.00	15.20	3.98	-	5.07	-

TABLE 2. - Chemical composition of ashes of different kinds of lignites and of cokes from true coal, percent (continued)

Origin	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O+Na ₂ O	S	P ₂ O ₅
<u>Coke from true coal:</u>								
11. Ruhr I	41.6	28.5	14.2	5.6	2.1	4.11	0.95	0.56
12. Ruhr II	49.9	29.4	13.4	2.3	2.3	-	0.28	1.08
13. Aix la Chapelle	42.2	28.1	14.8	3.9	2.7	5.1	0.87	0.50
14. Upper Silesia	24.4	14.6	20.1	18.2	9.7	-	3.45	0.16
15. Ostrau	37.2	25.2	13.3	10.9	4.7	2.3	1.40	2.19
16. Ruhr III	36.8	30.9	17.1	5.8	6.5	-	1.00	0.54
17. Ruhr IV	33.1	30.1	23.1	10.4	8.8	-	3.20	0.45
18. Ruhr V	35.0	26.9	14.3	9.8	3.1	-	1.92	0.62

Contents of Sulfur

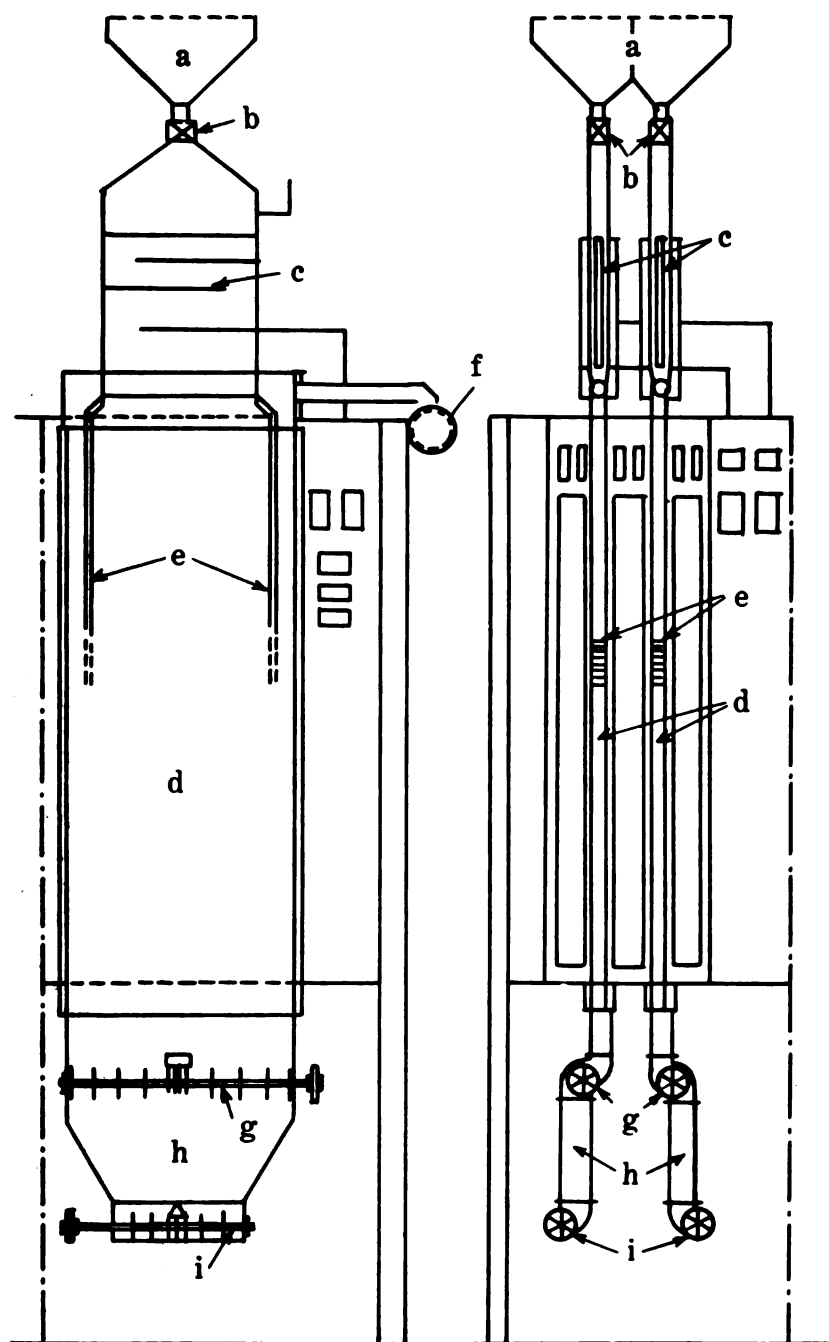
On account of the unfavorable properties of pig iron, due to the presence of sulfur, and considering the amount of work the blast furnace has to perform in order to reduce the contents of sulfur in the pig iron to a minimum, it is obvious that in the choice of basic fuels for the manufacture of coke those with low contents of sulfur deserve preference. During carbonization of the fuel the sulfur is distributed over gas, tar, water, and coke, and it can be taken for granted that the contents of sulfur in the coke correspond to those of the original fuel. As far as conditions permit, one will endeavor to produce a lignite coke with a sulfur content approaching that of normal coke, equal to about 0.8 to 1.2 percent. There is, however, the possibility, if operating with a basic slag also from a burden which, in relation to 1 ton of metal, contains up to 40 kilos of sulfur, inclusive of the sulfur contained in the lignite coke, to produce a pig iron with a sulfur content not exceeding 0.1 percent. For such an operation comparatively rich ores are required so as to limit the amounts of slag produced. Otherwise, the sulfur-removing process by means of soda is being applied, which, even if the contents of sulfur is as high as 0.7 percent, will effect a considerable reduction of the contents of sulfur.

By such an after-treatment of the pig iron with moderate silicon content and the application of a mixture consisting of soda, limestone, and fluorspar, or using soda only, reducing the contents of sulfur to 0.1 percent in a two-stage process, the following amounts of soda will be consumed:

Percent of sulfur in pig iron	0.7	0.5	0.2
Consumption of soda per ton of pig iron, kilos	25	20	10

In the first stage, the contents of sulfur are reduced to 0.2 percent and in the second to 0.1 percent.

The question as to how far the contents of sulfur are to be reduced inside the blast furnace and what portion is to be subjected to the after-treatment in the ladle can only be answered in consideration of the conditions of the ore and is further dependent upon the costs for coke, lime, and soda.



- | | | |
|----------------|--------------------------|-------------------------|
| a. Coal bunker | d. Carbonization chamber | g. Coke extractor |
| b. Cell valve | e. Baffle wells | h. Coke-cooling chamber |
| c. Predrier | f. Collecting main | i. Coke-discharge valve |

Figure 3. - Uniflow chamber oven in two vertical sections.

Manufacture of Coke from Lignites

The manufacture of a suitable blast-furnace coke from lignite is accomplished in several stages, which will be outlined briefly in the following. The lignite is carbonized in vertical chambers heated from the outside and constructed for continuous operations. Their particular construction effects forcible passage of the gases expelled within the chamber, and, owing to this peculiarity, the process is termed "uniflow" process. The unique construction of this oven, which has been protected by several patents, rests principally upon the fact that the primary liberated vaporous hydrocarbons are subjected to an influenceable thermic decomposition. This is characterized by a changed combination of the molecules of the hydrocarbons, and under these conditions byproducts are being formed which, in respect to their chemical composition as well as to the height of the yield, correspond to those recovered by the high-temperature carbonization of true coal, such as (1) crude benzole, (2) aromatic coal tar, and (3) ammonia.

At the same time, a coke is being produced which, in respect to its physical properties, is particularly suited for conversion into a metallurgical coke.

The Uniflow chamber oven. - The continuously operated chamber oven developed for this purpose is shown diagrammatically in two vertical sections in figure 3. The fuel slides from the bunker a, through the cell valve b, into the drier c, in which not only the water evaporates but also a certain preheating of the fuel is effected. From the drier c, the fuel slides down into the carbonizing chamber d, which is constructed of brickwork operated by high temperatures and heated from the outside. In this the fuel is, depending on its composition, subjected to the most favorable temperature and carbonized. By the walls e, suspended in the upper part of the chamber, the gases and vapors expelled from the fuel are compelled to travel for a certain distance in the same direction as the downward-sliding charge. Under the influence of the hot fuel and brickwork surfaces, the already mentioned molecular charges of the hydrocarbons are effected. The gas is taken off from the spaces formed behind the suspended walls e, and, passing through a collecting main f, is conducted through a cooling, tar-extracting, and scrubbing plant of usual construction to recover the byproducts. The coke is discharged continually on the base of the chambers by a mechanical extractor in the shape of a star roller g, the revolutions of which can be adjusted within wide limits. The coke is being cooled before extraction inside of the chamber h, and by means of a discharge valve i is temporarily withdrawn onto a conveying arrangement. The yield and the property of the products will be dealt with briefly hereinafter, and it may be mentioned in this connection that the height of the yield is mainly dependent on the chemical composition of the fuel carbonized. The adjustment of the temperatures of the oven and of the speed of throughput of the charge facilitate the manufacture of a coke which, in respect to its contents of volatile matter and on account of other properties, is particularly suited for transforming into a metallurgical coke.

Gas. - If lignites are subjected to low temperature carbonization, the yield of gas ranges between 100 and 150 m³ per ton, whereas with this process it is considerably higher on account of the additional formation of gas due

to the thermic decomposition of the tar vapors; further, a part of the moisture introduced with the charge participates in the formation of gas. Depending on the quality of the basic fuel and on the adjustment of the operation, 380 to 480 m³ gas is produced per ton of throughput. Correspondingly, the composition of the gas varies within about the following limits:

Composition of lignite gas

	<u>Percent</u>
Carbon dioxide	-8
Unsaturated hydrocarbons	2.5-1
Carbon monoxide	20-30
Hydrogen	45-50
Methane	15-8
Nitrogen	2.5-3
Gross calorific value	$\left\{ \begin{array}{l} 1/ 3860-3375 \\ 2/ 432-378 \end{array} \right.$
<u>1/ Calories per m³.</u>	
<u>2/ B.t.u. per cu. ft.</u>	

Tar. - The total yield of high-temperature tar plus the benzol absorbed from the gas corresponds to about 50 percent of the amount determined by analysis with the aluminum retort by F. Fischer, of which about 25 percent is crude benzol and 75 percent tar. The tar, as already indicated by its odor, has pronounced aromatic properties, and its specific gravity is about 1. Its content of phenols equals about 25 to 30 percent. The properties of the tar can be influenced within wide limits by corresponding adjustments of the operation, as indicated by the curve of the graph shown in figure 4, which accounts for two different kinds of tar produced from the same fuel by this process. The residual pitch remaining after distilling up to 340° C. amounts to 25 to 45 percent and is thus considerably less than from normal coal tar. The proportion of oils is correspondingly larger, thus affecting the economical results of the process very favorably.

Crude benzol. - Depending on adjustments of operating the ovens also, the quality of the benzol can be influenced within certain limits. The course of the range of boiling points, according to the distillation test by Engler, is reproduced by the curve shown in figure 5, whereas the chemical and physical properties correspond to the following values:

Properties of benzol from the carbonization of lignite

Specific gravity at 20° C.	0.8-0.85
Boiling range number	95-105
Loss in washing and distilling, percent	10-6
Composition, percent:	
Olefines	24-20
Aromatics	68-78
Paraffines and naphthenes	9-2

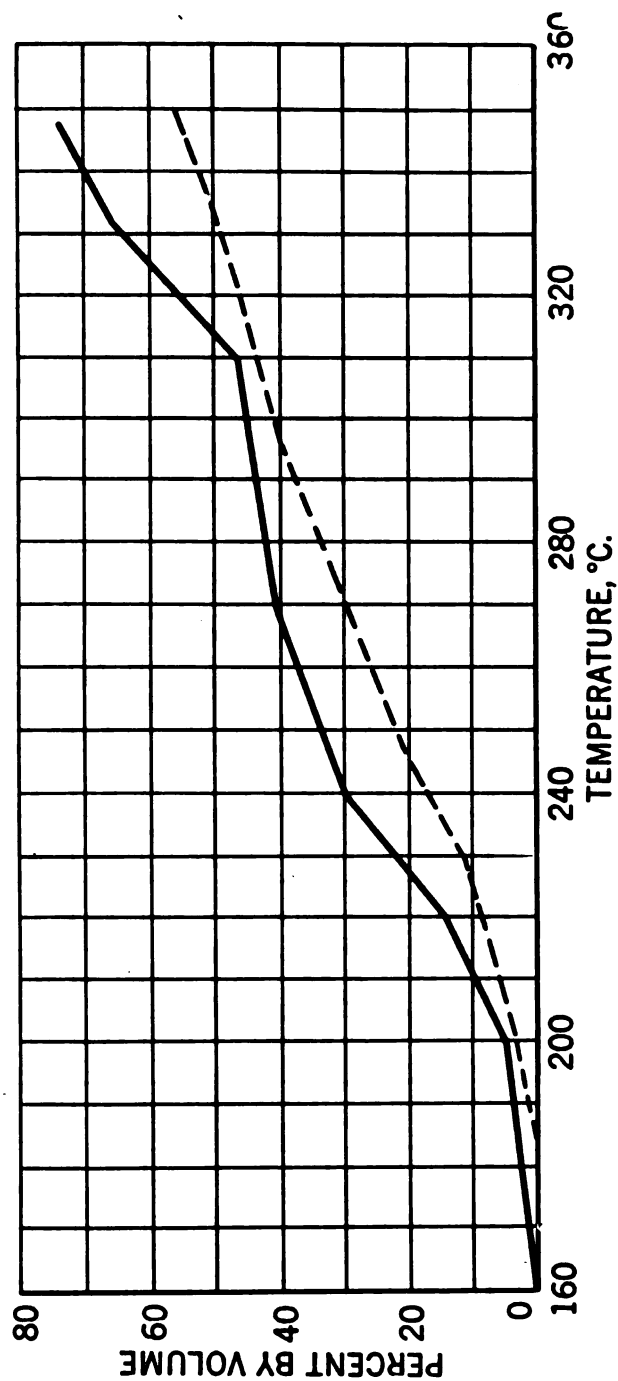


Figure 4. - Distillation curves of two tars recovered from the same lignite.

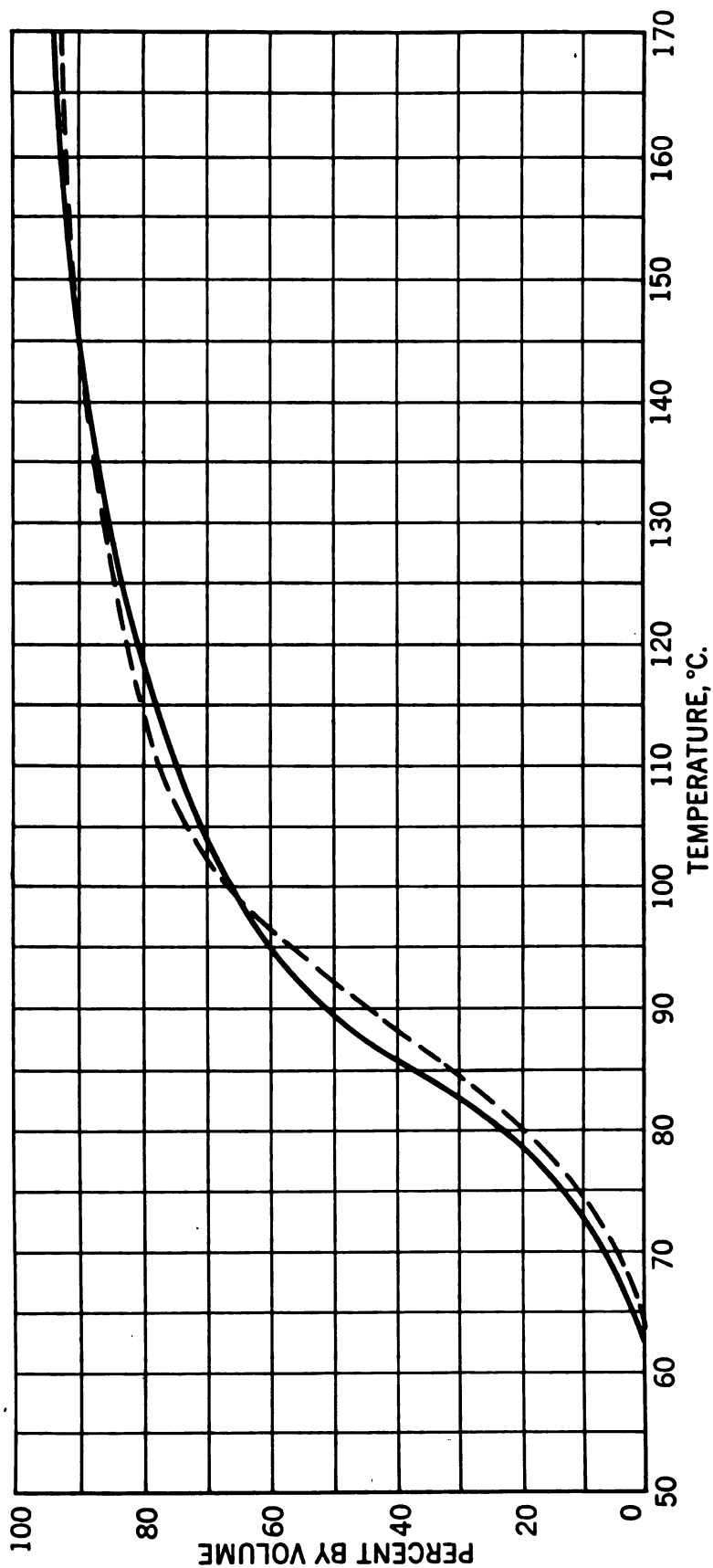


Figure 5. - Distillation curve of benzol from lignite (Engler test).

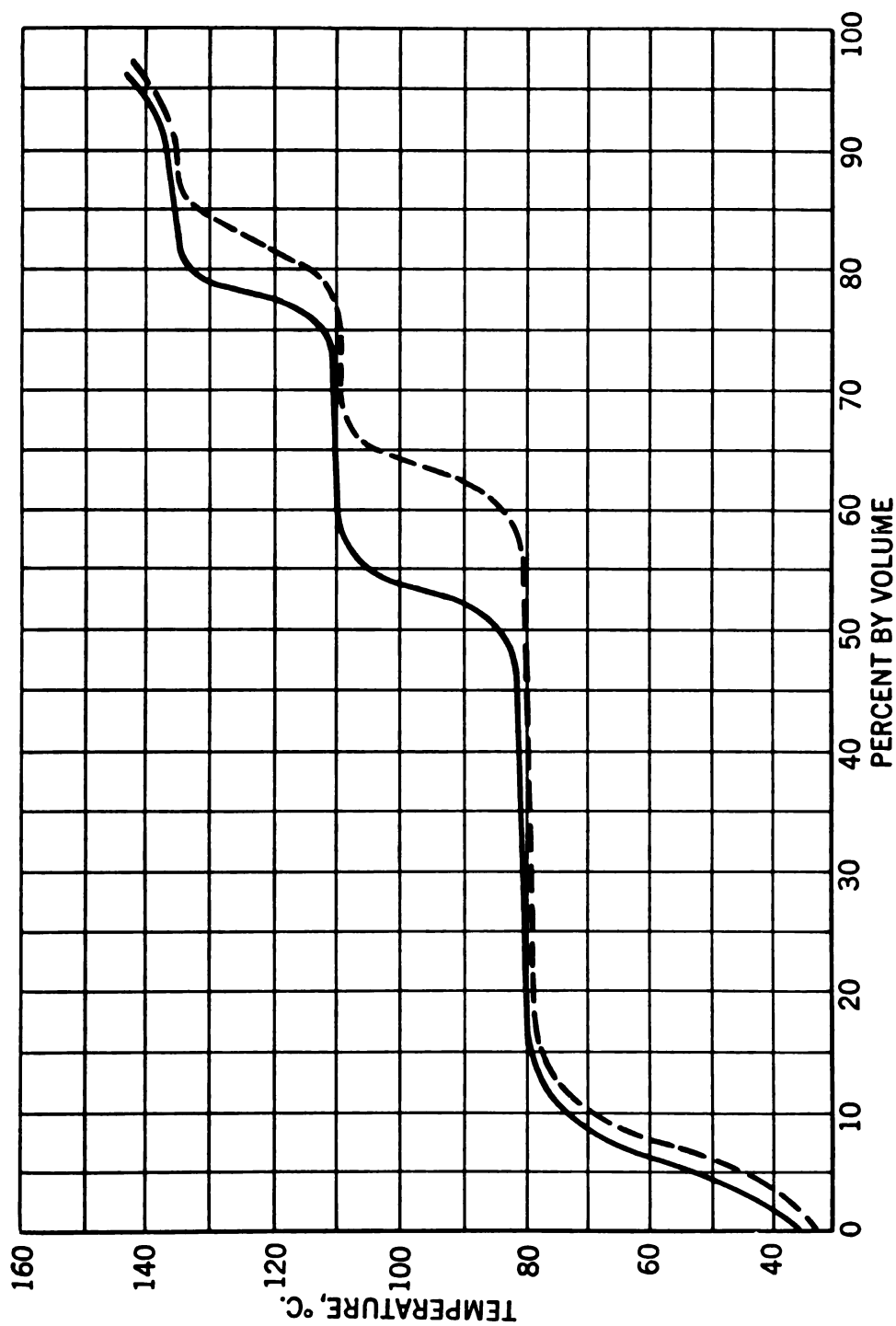


Figure 6. - Fine distillation curve of benzol from lignite.

The property of the benzol recovered in carbonizing lignite is further indicated by the two curves plotted in figure 6, which represent the course of the fine distillation and from which can be readily deduced that the boiling limits of benzol, toluol, and xylol are characterized by distinct stopping points. These curves also show that the quantity of each single fraction can be varied by a corresponding adjustment of the operating conditions.

The properties of the respective fractions of benzol, toluol, and xylol correspond in every respect with those recovered from crude benzol absorbed from coke-oven gas, so that further working up can be effected in different ways, as, for instance, (1) to pure compounds like benzol, toluol, etc., by fractionating, or (2) to motor benzol.

The benzol thus produced has excellent properties as a motor fuel, and already the crude product tested in the C.F.R. motor had octane figures ranging from 95 to 105. With the motor benzol, these figures come out still more favorable, and it is free from constituents that attack metals.

Coke. - The yield of coke depends directly on the properties and composition of the basic fuel. In practical operation, the yield is about 1.5 to 3 percent higher, compared with the results of the crucible test. The contents of volatile matter in the coke which, just as the contents of moisture, are of particular importance for the further transforming into metallurgical coke, can, as already mentioned, be maintained easily at the desired rate by corresponding adjustments of the continuous operation of the ovens. As the coke recovered from the first stage of the process represents an intermediate product only, which is to be worked up further, its properties need not be taken further into account here.

Briquetting. - The coke recovered in the first stage of this process is, subjected to briquetting to transform it into a metallurgical coke suitable as a blast-furnace fuel. This is accomplished by, first of all, crushing the cooled coke in a crusher of special construction to a grain size so as to assure its most favorable physical condition. The crushed coke is subsequently mixed with caking coal and a suitable binding medium, and depending on the properties of the coke, mostly coal-tar pitch recovered as a residual from the tar distillation, is used for this purpose. The proportion of pitch yielded by distilling the tar can be increased artificially by oxidizing and polymerization of the tar where circumstances demand such a course. Other additions and binding media, such as concentrated sulfide lye, higher alcohol, tar oils containing phenols, soft pitch, caking coal, etc., can, according to circumstances, be used for the same purpose.

By close research in this connection, extensive experience has proved that from virtually every kind of coke produced by this process a mixture suitable for subsequent briquetting can be prepared. It should be pointed out, because it is of particular importance, that the proportion of the binding medium to be added can remain comparatively small, so that the economical results of the process are affected but slightly from this source.

The kind of crushing and mixing, as well as the desired shape and weight of the individual briquettes, as well as the pressure applied during briquetting form the basis for the most favorable properties of the briquettes, and these conditions are supplemented by the subsequent thermic hardening of the briquettes, which forms the second stage of this process. In respect to the technic of this process, briquetting and thermic hardening form one single stage, as the binding medium applied is partly recovered by the hardening process and the pitch is again being used. This is of great importance for the economical results of the whole process.

Thermic hardening of coke briquettes. - The briquettes manufactured as indicated above are subsequently transferred to a second chamber-oven plant, and it is of importance to take into account their physical properties, beginning at the discharge of the semicoke from the first stage, its rate of cooling, and its crushing as well as its briquetting up to the point where the briquettes are again subjected to a thermic treatment, since the properties of the final shaped coke depend to a large extent upon the most favorable conditions of each operating phase.

The briquettes are hardened by carbonization, at a temperature of 900° to 1,000° C. This process commences with the evaporation of the light-boiling constituents of the binding medium and continues as carbonization of the residual matter, which effects a binding and cementing of the coal particles to each other, at the same time forming an evenly built up coke structure. The thermic decompositions of the binding medium accompanying carbonization under the temperatures stated effect formation of a coke skeleton that very closely resembles the normal coke oven coke in respect to its cell structure. Compared with the macrostructure of the normal coke, the shaped lignite coke manufactured by this process shows a very close-celled microstructure, which is responsible for the high reactivity of this fuel. The metallurgical lignite coke produced in this way is characterized by the following properties, which assure its application as a blast-furnace fuel:

Crushing resistance .. 200-300 kg/cm²
 Abrasive hardness 75-85 percent
 Ignition point 430-460° C.
 Reactivity at 900° C.. 70-80 percent

In this connection it should be mentioned that the yield of briquettes having the properties just stated is 90 to 95 percent, while 5 to 10 percent will not fully stand the tests, though this in no way impairs the value and the utilization of the product in total bulk.

The volumes of gas liberated during the thermic hardening treatment are introduced into the gas circuit of the plant after the evaporated pitch has been recovered by separation for further use.

Yields in relation to the calorific power of the fuel. - To provide a scale for judging the yields as well as the consumption of heat in the manufacture of metallurgical coke from lignite, the process will set in comparison to the production of coke from true coal in coke ovens. For this purpose the average values referring to modern coking are plotted in the shape of a

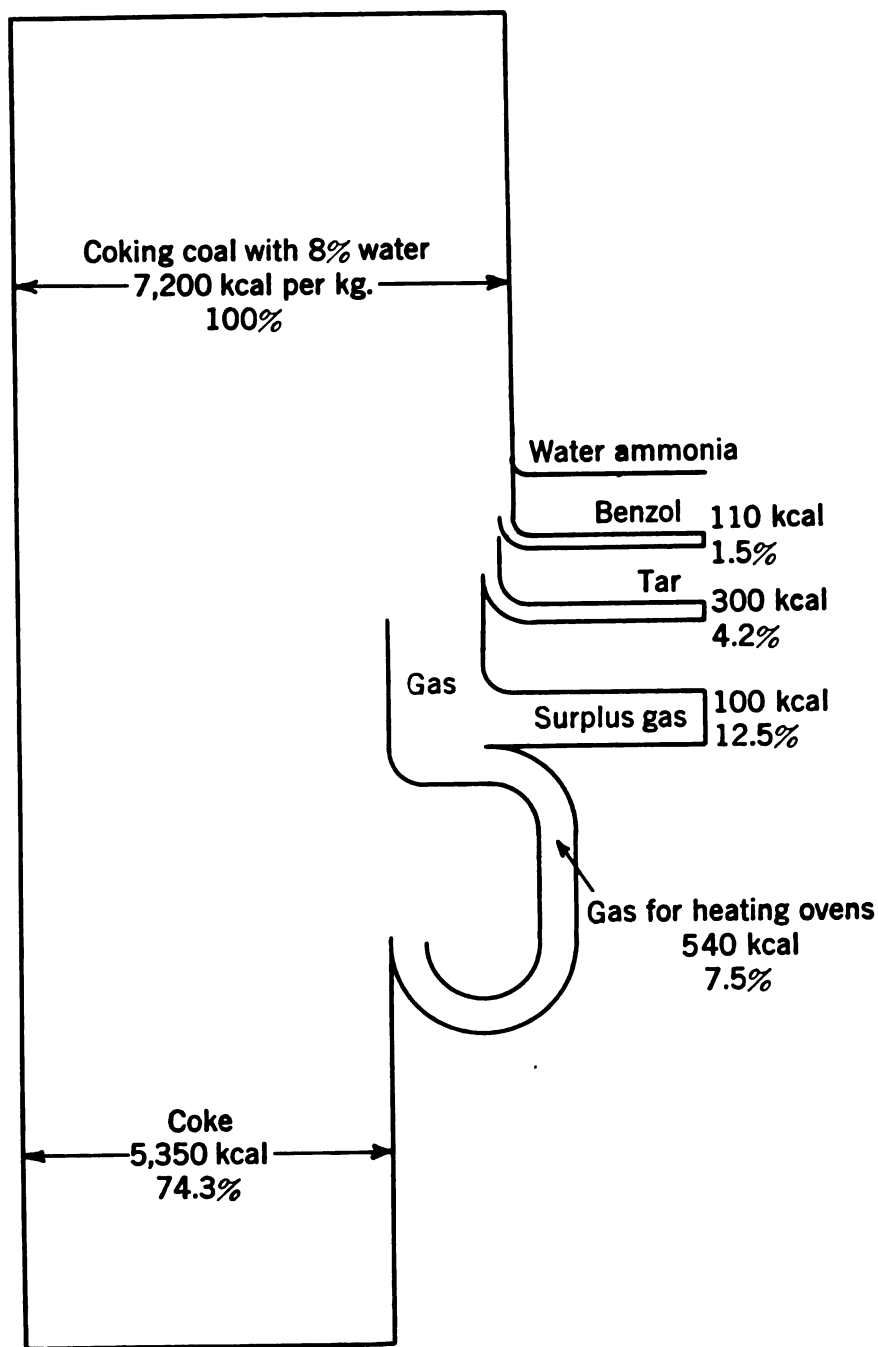


Figure 7. - Diagram of heat balance of modern coke oven.

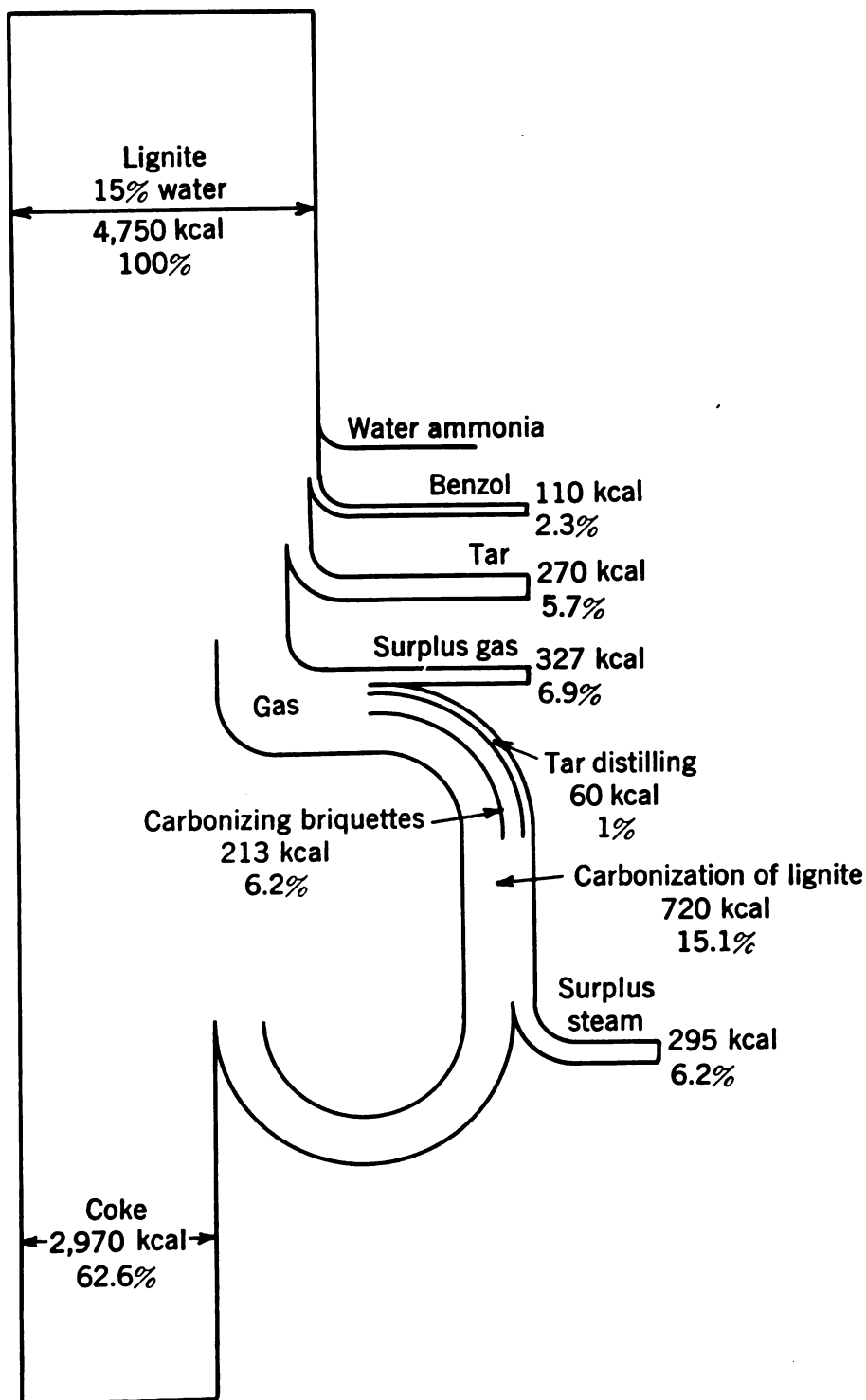


Figure 8. - Diagram of heat balance of two-stage process operating on lignite.

Sankey diagram shown in figure 7, starting with a coking coal containing 8 percent of moisture and having a gross calorific value of 7,200 kcal/kg. (1,296 B.t.u./lb.). The yield of coke is equal to 74.3 percent and the gross calorific value of the coke amounts to 5,350 kcal/kg. (642 B.t.u./lb.). All other yields as well as the consumption of heat can be taken from the diagram of the figure 7.

The diagram of the lignite carbonization is shown in figure 8 and first of all will be apparent the considerably lower potential heat of the lignite, amounting to 4,750 kcal/kg. (560 B.t.u./lb.), compared with the calorific value of the true coal as stated above. This affects principally the yield of coke, which amounts to only 62.6 percent of the lignite, compared with 74.3 percent of the true coal. The yields of the liquid products, however, like benzol and tar, equal to 2.3 and 5.7 percent, are much better in connection with the carbonization of lignite in comparison to the true coal, where they are equal to 1.5 and 4.2 percent only, in both cases, relative to the potential heat of the fuel charged. The further values can be taken so easily from the diagram of the figure 8 that they need not be considered further here.

Blast-furnace trial with shaped lignite coke. - A shaped coke produced from lignite by the two-stage process, as outlined in the foregoing, was tried on a normal blast furnace that had a shaft of 2.4 inside diameter to confirm its adaptability for this purpose. The other parts of the blast-furnace plant, as well as the condition of the furnace itself, did not offer very favorable circumstances for carrying out such a trial. Apart from the properties already mentioned, the lignite coke had the following composition:

	Percent
Water	2-4
Ash	18-20
Fixed carbon	97-93
Volatile matter	1-3
Sulfur	1.5-2

The ash had the following chemical composition:

Chemical composition of ash in lignite coke

Constituent	Determined as -	Percent
Silicon	SiO ₂	32.60
Iron	Fe ₂ O ₃	18.90
Alumina	Al ₂ O ₃	14.80
Lime	CaO	15.80
Magnesia	MgO	3.96
Sulfur	SO ₃	14.31
Rest	-	.37

An operation of the blast furnace requiring several days and using this shaped lignite coke exclusively proved that in spite of the high contents of ash and sulfur an excellent pig iron was made from a burden consisting of a mixture of hematite bog ore, roast spar, and brown ore. The composition of the pig iron produced during this trial run corresponds to the following:

Composition of pig iron

Constituent	Formula	Percent
Manganese	Mn	2.090
Phosphorous	P	0.046
Copper	Cu	0.117
Antimony	Sb	0.052
Carbon	C	2.470
Sulfur	S	0.170
Silica	Si	0.414
Iron	Fe	94.041

The consumption of coke in the blast furnace in relation to 1 ton of pig iron increased in comparison to the coke normally used in direct proportion to the higher contents of ash in the lignite coke.

From the results of this trial, carried out on a commercial scale, could be deduced that the shaped lignite coke manufactured by the two-stage process meets the conditions of normal blast-furnace operation in every respect.

Economy. - The manufacture of a metallurgical-shaped coke from lignite by the two-stage process is absolutely economical, particularly if production costs are compared with coke-oven coke, which must be imported from distant places and must thus be debited with high freights.

In a particular case in which a lignite served as basic fuel and contained so much water that it had to be subjected to predrying, the price of the lignite amounted to 8.50 M. per ton after its content of water was reduced to 15 to 16 percent by drying. The costs of the shaped coke manufactured by the two-stage process at a plant having an output of 300 tons of coke per day amounted to 19.20 M. per ton of coke under the local conditions given. In judging these costs, it must be taken into account that the artificial drying of the lignite when its high content of water increased the manufacturing costs very heavily and by a basic fuel which in the raw state does not contain more than 15 to 16 percent water, the coke probably could be supplied at a much lower price, causing naturally a correspondingly large reduction in the manufacturing costs of the coke.

True coal. - The efforts made to increase the yield of tar from coal by subjecting it to low-temperature carbonization are well known. The further development of these processes was held up by the impossibility of producing a hard coke that would withstand abrasion. By the necessity of producing as much tar as possible, in addition to a good hard coke, a new problem had to be faced, the solution of which is of the utmost importance for these coal deposits, which consist of geologically young or noncoking kinds, but which, on account of their high contents of tar, are very suitable for treatment by the low-temperature carbonization process. The semicoke produced by these processes can, because of its soft and brittle nature, be classed only as a minor fuel. The problem indicated was the recovery of a hard metallurgical coke with good abrasive properties instead of the inferior semicoke. The solution of this problem is also, for low-temperature carbonization on its

broadest base, of the utmost importance. As the market for a good metallurgical coke is almost unlimited and is still increasing, it can readily be assumed that it should be possible to accelerate the development of low-temperature carbonization on the basis of true coal in a much larger measure than was ever contemplated in order to recover a good metallurgical coke and in addition large amounts of valuable tar oils.

Compared with the high- and low-temperature carbonization processes applied so far, by which the coal is subjected to a thermic treatment, it is suggested that the coal be worked up by the two-stage process, by which the weakly-caking or noncaking coal with a high content of volatile matter is transformed into semicoke in the first stage at the same time gaining a maximum yield of tar. In the second stage, the semicoke is worked up to a high-class metallurgical coke of even shape by thermic treatment. For this purpose, briquettes are being made from the semicoke exactly as coal is usually transformed into briquettes. As a binding medium for briquetting, hard or soft coal-tar pitch as well as caking coal are available. The briquettes are being formed at a pressure of 100 to 200 atmospheres in a roller press of special construction. The raw briquettes thus produced are subsequently carbonized at 900° to 1,000° C., retain their shape completely, and yield a very hard and dense coke with excellent abrasive properties. For the final thermic treatment, vertical chamber ovens are applied, which may be constructed for continuous or intermittent operation.

TREATMENT OF COAL BY THE TWO-STAGE PROCESS

The application of the two-stage process for the manufacture of shaped metallurgical coke from coal is comparatively simple. The independence of the process from the physical properties of the basic coal is also a particular advantage in this instance. A deficiency of agglutinating power is very favorable for the treatment of true coal, and it should be mentioned here that coals with high contents of tar and having no agglutinating power at all hardly exist. In this case a certain yield of tar is also very desirable to improve the economical results and at the same time the tar forms the source for the coal-tar pitch that serves as a binding medium for manufacturing the briquettes from the semicoke produced in the first stage.

First Stage of Process

The contents of water in all coal are so low that without requiring artificial drying it can be directly carbonized, crushed to a suitable size. As an exception may be mentioned the slimes recovered as residual coal from the settling ponds of coal washeries or the fine coal that has been treated by a froth flotation process. Such kinds must naturally be subjected to artificial drying before carbonization.

In place of the uniflow chamber operating at high temperatures and mentioned above for the carbonization of lignites, the thermic treatment of coal is effected in the first stage by low-temperature carbonization ovens in order to obtain a maximum yield of tar and a high reactivity of the semicoke. Ceramic low-temperature carbonization ovens have been developed especially for this purpose and have given every satisfaction in operation on a commercial scale.

The ceramic low-temperature carbonization ovens of the Didier-Werke differ from the construction shown in figure 3 on the whole only in that the suspended baffle walls in the top of the carbonization chambers are missing, and by the mode of heating, which is arranged so that a comparatively low temperature is evenly distributed over the whole wall surface on each side of the chamber. A further arrangement must be mentioned which has not been applied to the ovens reproduced in the figure 3, consisting of a horizontal gas-distributing cylinder placed above the coke extractor roller *g* to one side of the chamber for the purpose of being able to introduce circulating gases on the base of each chamber equally distributed over the whole sectional area. These gases ascend through the charge and impart their sensible heat to the coal particles to improve the transfer of the heat of the oven, and serve also as a means to adjust the time during which the gases and vapors evolved from the coal are allowed to remain inside the chamber.

In the first stage of the process, by the thermic treatment of coal, semicoke, low-temperature tar, light oil, and a gas of high calorific value are produced while the gas remains to a certain extent in circulation. The yield of low-temperature tar, light oil, and gas depends not only on the temperature of the oven but, apart from the quality of the coal treated, to a large extent on the time which the gases and vapors evolved are allowed to remain inside of the chamber. This length of time is adjusted by addition of larger or smaller volumes of circulating gas, and this adjustment shows an almost instantaneous effect. As a circulating gas, the gas evolved from the coal is used after it has been freed from tar, and a portion of this is branched off and conducted through the chambers. The low temperature carbonization oven of the Didier-Werke is suitable for the thermic treatment of noncoking coals having a grain size between 3 and 50 mm., as this provides the most favorable conditions for the operation of the automatic coke extractor. Larger proportions of smalls in the charge impair the even discharge of the coke and are responsible for the presence of dust in the tar, the value of which is reduced thereby, particularly also because a certain percentage of dust effectively prevents its dehydration. Coal pieces exceeding 60 mm. in size are not favorable for its low-temperature carbonization, because the heat does not penetrate fast enough to the center of the individual pieces and causes tarry zones to remain, which disturb the even quality of the semicoke while the smaller pieces have been subjected to the effects of the heat for too long a time.

If the problem is to be dealt with to produce a metallurgical coke in even shapes from powdered coal, flotated coal, coal dust, or coal slime, another type of low-temperature carbonization oven adapted to the physical condition of such materials will be applied.

The turbine drier of the Buttner-Werke shown in figure 1 has in the meantime, by retaining its basic constructional details, been adapted for the low-temperature carbonization of fine-grained or powdery noncoking coals and has proved to have a comparatively high throughput without causing the development of unduly large amounts of dust.

Briquetting

The briquetting of the semicoke produced in the low-temperature ovens of the first stage of the process is effected in the same way as the manufacture of coal briquettes. The semicoke is, first of all, crushed, as this is necessary to obtain an even cell structure in the final coke. The crushed semicoke is intensively mixed with about 10 percent of good caking coal and about 6 percent of coal tar pitch. A further agitating by the application of heat from indirect steam raises the temperature of the mixture to about 105° C., and in this state it is transferred to roller presses, in which, under a pressure of 100 to 200 atmospheres, ovoids or briquettes of desired shape are produced.

The economy of the process depends largely on the quantity of coal-tar pitch used as a binding medium for the briquetting, and as the semicoke of the Carbocoal process mentioned above required as much as 12 percent of coal-tar pitch for its briquetting, the economic results of the whole process were also, for this reason, very much impaired. By the present process, the proportion of pitch as a binder added to the semicoke is thus kept at a level that just suffices for the formation of an evenly dense structure of the coke. In addition, part of the coal-tar pitch is substituted by caking coal, which by itself suffices, if added in adequate proportion, to produce a close and even structure of the hard shaped coke.

During carbonization of the raw briquettes in the second stage of the process, the low-boiling parts of the coal-tar pitch are expelled and subsequently recovered while the gas is being cooled. Since at the same time, also, the tar is being formed by the carbonization of the caking coal added to the semicoke for briquetting, the tar recovered remains sufficiently liquid so that it does not cause any obstructions. The soft pitch recovered is again applied as a binding medium for briquetting, so that by adding 6 percent of pitch, 3 percent of this can be substituted by the soft pitch recovered in the second stage. From these facts, it can be readily deduced that the coal-tar pitch recovered from the low-temperature tar of the first stage, together with the soft pitch recovered in the second stage, plus the coking coal also serving as a binding medium, suffice completely to cover the requirements of binding agents of this process.

It has already been mentioned that also the semicoke made from lignite can be briquetted by using pitch as a binder and can subsequently be transformed, by a thermic treatment, to hard coke briquettes. To accomplish this, addition of caking coal to the semicoke is an absolute necessity, not so much for saving binding agents as to effect the formation of a dense and even structure of the briquetted coke, as without this addition a dense coke structure is not obtainable from lignite semicoke. The semicoke from coal has, in comparison to the semicoke from lignite, not a capillary structure, and the addition of caking coal to the former simply serves to save pitch so as to improve the economical results of the process. By the manufacture of shaped coke from lignite semicoke, the caking coal, together with the pitch, forms a preliminary assumption to convert the small-grained semicoke into a coherent, dense, and hard shaped coke.

Second Stage of Process

The briquettes manufactured, by the addition of a binder, from the semi-coke of the first stage of the process are hardened in the ovens of the second stage by a thermic treatment. For this purpose, vertical-chamber ovens of standard construction are applied, which are designed for continuous operation. Also, intermittently working vertical chamber ovens can be applied for this purpose, but preference is given to the continuously operated type on account of the more gentle treatment of the briquettes and also because the continuously operated chamber oven assures better utilization of the heat. As the intermittently operated ovens must be discharged completely each time a pretty high initial drop of the briquettes in the beginning of the charging operation cannot be avoided, by which the briquettes will naturally suffer, while the continuously operated chambers remain filled and the drop of the charge corresponds to the even discharge of the coke on the base. The chambers are heated, so that the briquettes are subjected to a temperature of 900° to 1,000° C., and during this time the lower-boiling parts of the pitch are expelled, together with the tar vapors of the caking coal added.

Compared with the normal operation of such vertical-chamber ovens in gasworks, the present construction allows, just as in connection with the low-temperature carbonizing, vertical chamber ovens of the first stage for the improvement of the transfer of the heat by having arrangements provided that permit the introduction of circulating gas freed from tar on the base of the chambers.

The coke produced in the form of carbonized briquettes has already been cooled down inside the oven chamber to about 200° C., so that its sensible heat is being recovered for the oven and quenching of the coke is hardly necessary.

Mode of Operation of Two-Stage Process

The diagrammatic table in figure 9 indicates the quantities of fuels required for the manufacture of 150 tons of shaped metallurgical coke per day. As a basis serve 200 tons of coal, which in the first stage yield 140 tons of semicoke and 18 to 20 tons of low-temperature tar. To the semicoke are added about 10 percent of caking coal and 6 percent of coal-tar pitch, but these quantities are not mentioned in figure 9, because their proportion is subject to alterations and dependent on the quality of the semicoke. The semicoke, together with the binding agents, is passed through the briquetting press, yielding about 170 tons of raw briquettes, from which, in the second stage of the process, a certain quantity of tar is being recovered, which is added to the low temperature tar of the first stage. The total tar is distilled to recover the residual pitch. As final products of the process, based upon a throughput of 200 tons of coal per day, thus appear 150 tons of shaped metallurgical coke and 13 to 15 tons of tar oils.

From the flow sheet represented by figure 10 can be deduced that for a throughput of 200 tons of coal per day, three low-temperature carbonization chambers are required in the first stage for the production of about 160 tons shaped metallurgical coke, both types of ovens in the first and in the second

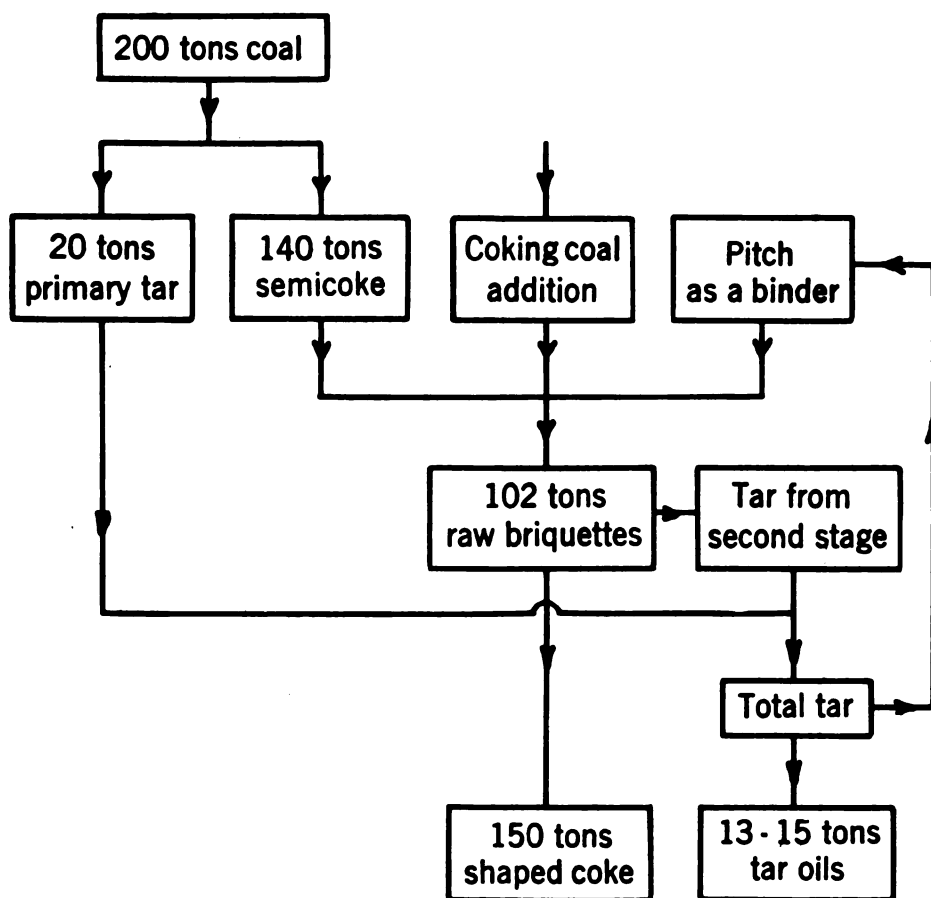


Figure 9. - Distribution of raw materials in two-stage process operating on true coal.

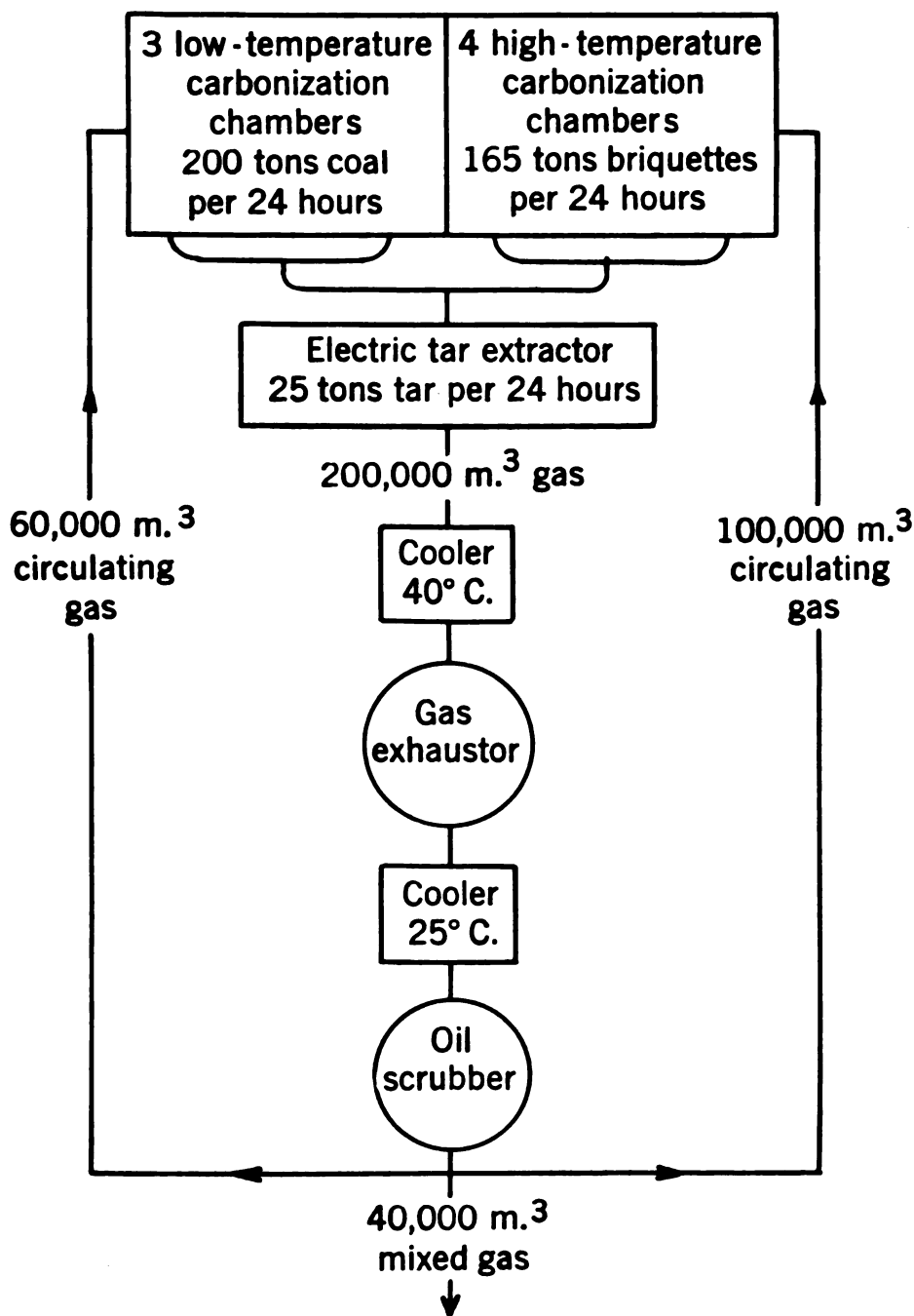


Figure 10. - Flow sheet of a two-stage plant operating on coal with a single gas offtake.

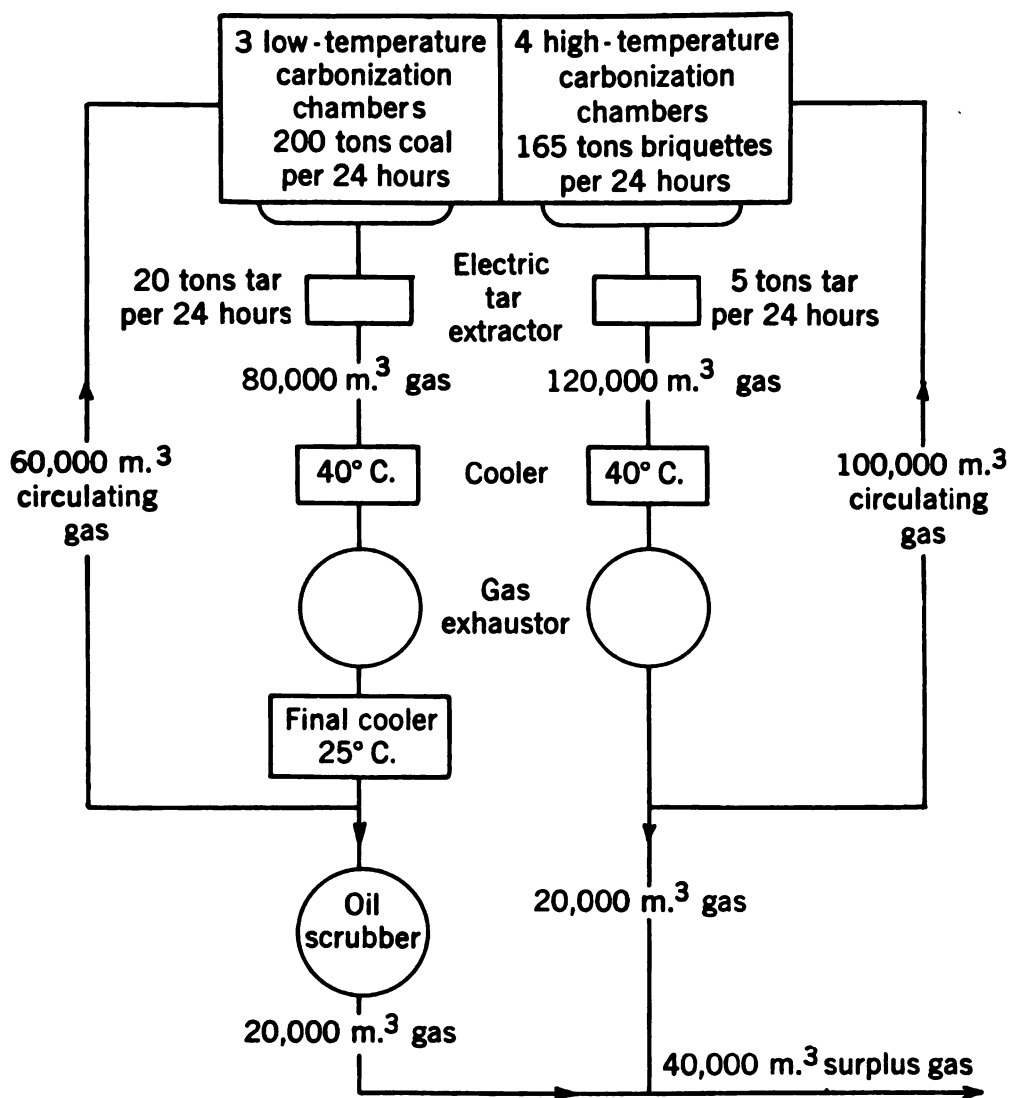


Figure 11. - Flow sheet of a two-stage plant operating on coal, with separate gas offtakes.

stage are heated by producer gas, and the raw gas entering the collecting mains of both types is drawn off by a common suction main and passed, first of all, through an electric tar extractor, in which a temperature well above the dew point of the gas is maintained. In this, about 25 tons of tar are recovered per day free from water. The gas, representing a volume of about 200,000 m³, is now subjected to cooling and enters the gas exhauster with a temperature of about 40° C. The gas is now conducted through the final coolers, lowering the gas temperature to 25° C., and in this condition it is passed through the oil scrubbers to absorb the light oil. The stripped gas is now divided, and 40,000 m³ mixed gas having a calorific value of 4,300 kcal/m³ (482 B.t.u./cbft) are at disposal for other purposes, while 60,000 m³ remain as circulating gas for the ovens of the first stage and 100,000 m³ for the carbonization ovens of the second stage.

The flow sheet shown in the figure 11, being based on a plant of the capacity mentioned above, indicates the tar extraction and the cooling of the gas in two separate gas-treating plants, one for the low-temperature carbonization ovens and one for the high-temperature ovens of the second stage, so that each stage is connected to a separate gas-treating plant, which unite on the end. From the ovens of the first stage, 80,000 m³ low-temperature coal gas are taken off per day and passed at a temperature above its dew point through the electric tar extractor, by which 20 tons of tar free from water are recovered. The gas is then cooled to a temperature of 40° C., and the gas exhaustor propels it through the final coolers, on the outlet of which the gas temperature has dropped to 25° C. To reduce the work performed by the oil scrubbers, 60,000 m³ gas per day are diverted immediately after the final coolers, serving as circulating gas for the ovens of the first stage. On the outlet of the oil scrubbers, 20,000 m³ gas per day having a gross calorific value of 5,500 kcal/m³ (616 B.t.u./cbft) are disposed of for suitable consumption elsewhere. The 120,000 m³ gas per day taken off from the carbonization ovens of the second stage are passed, at a temperature above its dew point, through the electric tar extractor, from which 5 tons of tar free from water and having high contents of soft pitch are recovered and its presence makes a separate treatment of the gas from the second stage of the process in many cases recommendable, as indicated in the figure 11. The gas freed from tar is cooled to about 40° C. and propelled further on by the gas exhauster, representing 100,000 m³ circulating gas per day. This is being passed through the carbonization ovens of the second stage and 20,000 m³ gas daily to be mixed with the gas from the ovens of the first stage. As the gas from the ovens of the second stage has a gross calorific value of 3,000 kcal/m³ (336 B.t.u./cbft), the mixture of the gases from both stages is equal to a calorific value of 4,300 kcal/m³ (516 B.t.u./cbft); its volume per day represents 40,000 m³ available as surplus gas.

Throughputs and Yields

For the results indicated in figure 9 to 11, based upon the treatment of coal from Upper Silesia, and with reference to the yields and throughputs of such a two-stage process plant, the following particulars are available:

Yields and Outputs Per Day

	Metric tons
Coal throughput per day	200
Semicoke yield	140
Coking coal added	14
Coal-tar pitch binder	8
Raw briquettes	362

Low-temperature tar (90 percent from 11 percent tar by test = 10 percent)	20
Soft pitch from second stage (50 percent of the pitch)	4.2
Tar from coking coal (70 percent from 8 percent tar by test	.8
Crude gasoline, 34 grammes per m ³ in 80,000 m ³	2.72
Crude gasoline, 2.5 grammes in 120,000 m ³	.30

The gasoline output is equal to 3.02 tons in 200,000 m³, corresponding to 15.1 grammes per cubic meter. With 3 grammes of gasoline remaining in the stripped gas, 2.42 tons, equal to 15.1 grammes per cubic meter, are recoverable per day.

Properties of Shaped Coke

As to the properties of the shaped coke manufactured by the two-stage process from true coal, a number of results are available which, as example of many trials, are stated in the following, representing in each case a good average.

Shaped coke from coal of Upper Silesia

<u>Proximate analysis</u>	<u>Percentage</u>
Water	3.2
Ash	13.8
Volatile matter	10.2
Fixed carbon	72.8
	100.0

Binding medium used: Coal-tar pitch with softening point at 69.5° C. recovered in the process.

Briquetting: Pressure applied: 200 atmospheres; temperature of briquetting mixture: 90° C.

Carbonization: Temperature in ovens of the second stage: 950° C.

The trials are based upon two different mixtures, which were composed as follows, differing from each other only by their size of grain.

Briquetting mixtures

	<u>Mixture I</u>		<u>Mixture II</u>	
	Grain size, mm.	Percent	Grain size, mm.	Percent
Semicoke	0-3	82	0-0.5	82
Caking coal	0-0.5	11	0-0.5	11
Coal-tar pitch	0-0.25	7	0-0.25	7

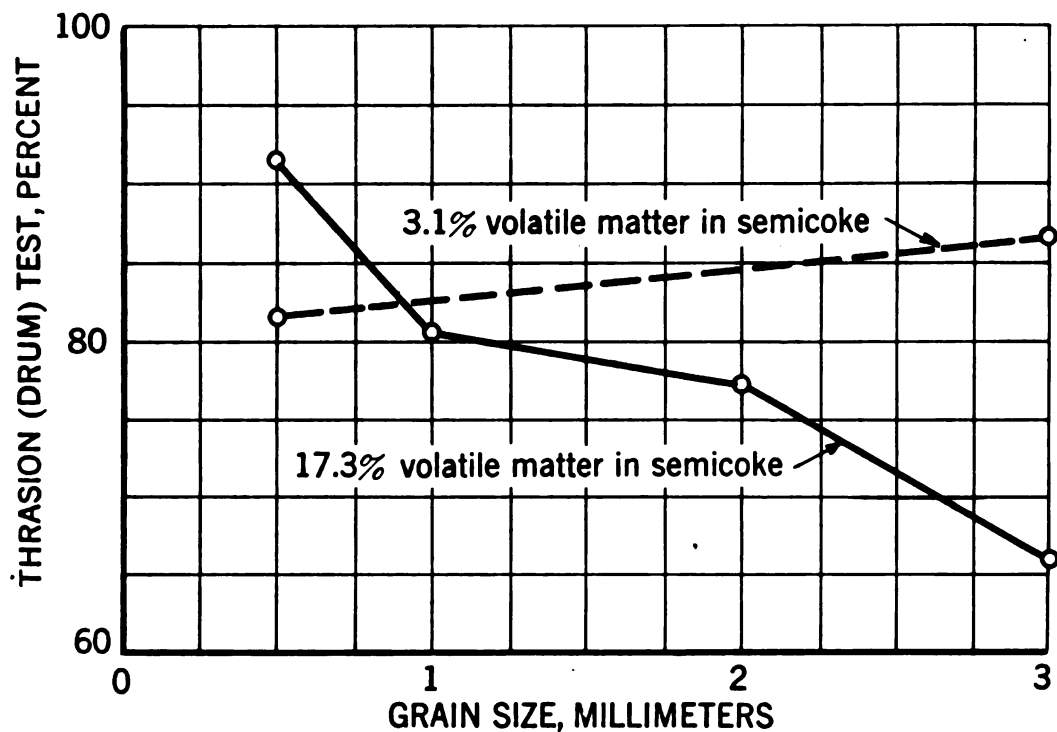


Figure 12. - Effect of grain size and volatile matter of semicoke on the abrasive resistance of the finished shaped coke.

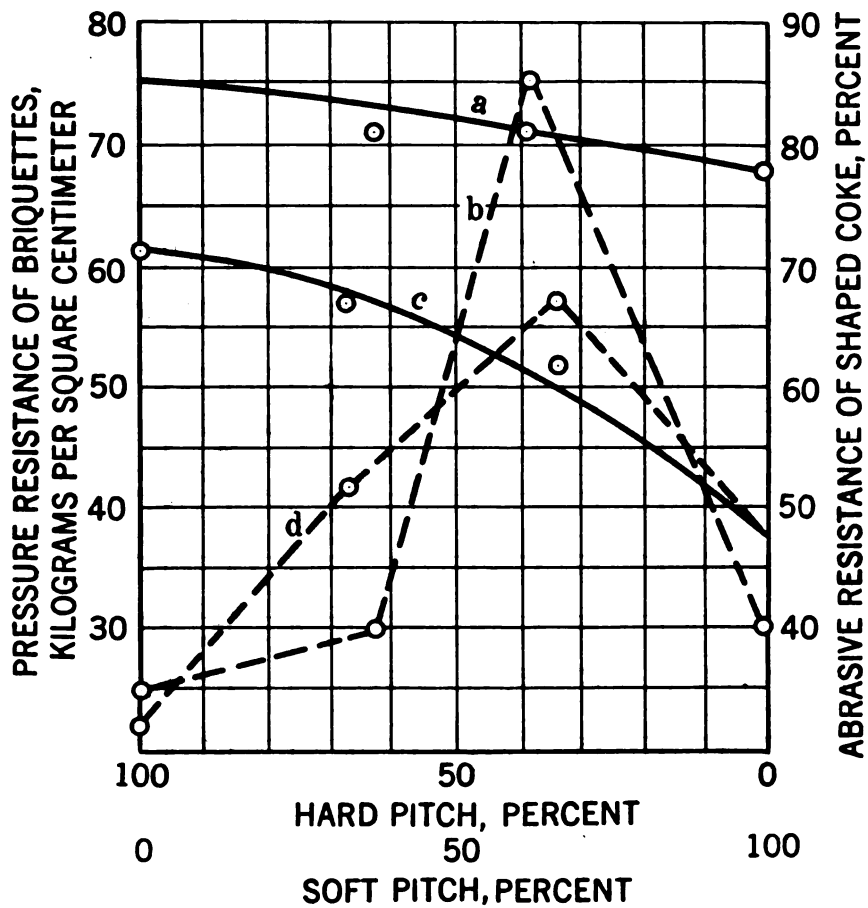


Figure 13. - Effect of binding medium of briquettes upon pressure and abrasive resistance of the finished shape coke.
 a = Abrasion resistance of briquettes with 8 percent pitch.
 b = Pressure resistance of briquettes with 8 percent pitch.
 c = Abrasion resistance of briquettes with 6 percent pitch.
 d = Pressure resistance of briquettes with 6 percent pitch.

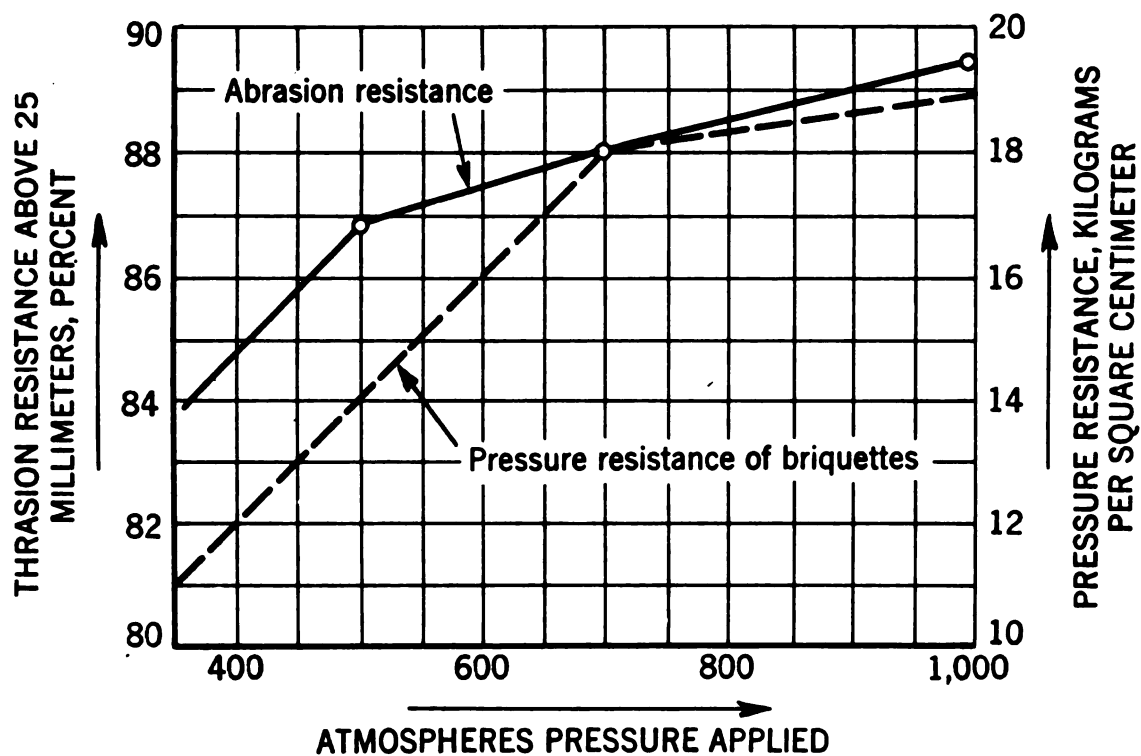


Figure 14. - Abrasive resistance of shaped coke made with caking coal only as binding medium.

Properties of shaped coke

Mixture	I	I	II	II
Pressure resistance, kg/cm ²	11.2	17.6	11.3	18.2
Contents of moisture, percent	3.1	8.3	2.9	7.3
Abrasion (drum) test, percent	83.4	86.8	88.4	93.0
Ignition point in oxygen, °C.	498	-	504	-

The results compiled above indicate that the abrasive properties of the briquettes improve with decreasing grain size of the briquetting mixture.

To produce a shaped metallurgical coke that must meet certain demands, exactly predetermined operating conditions must be maintained, which, apart from the thermic treatment of both stages of the process, also concern the selection of the most favorable grain size of the semicoke to be briquetted and, further, the quantity and quality of the binding medium as well as of the caking coal to be added, and eventually the briquetting conditions, which are all of great importance for the quality of the shaped coke produced. This whole range which sounds rather more extensive than it is in reality has in the mean time been fully elucidated in all directions by intensive research work, so that all individual details independent from the properties of the basic coal can be mastered readily and under these conditions the manufacture of a hard shaped metallurgical coke does not present any difficulties whatever.

Figure 12 represents a graph from which the effect of the grain size of the semicoke and its contents of volatile matter upon the abrasive properties of the shaped coke can be deduced. The briquetting mixture consisted of 85 percent semicoke, 9 percent caking coal, both having a grain size of 0 to 0.5 mm., and 6 percent of pitch were added as a binder. The straight running curve representing the results of the abrasion test of the shaped coke is based upon a semicoke with 3.1 percent volatile matter. The course of these curves is affected by the grain size of the semicoke indicated by the vertical ordinates of the graph.

The graph shown in figure 13 indicates the effect of the binding medium upon the pressure and abrasion resistance of the shaped coke and, as shown by the vertical ordinates, the coal-tar pitch has been substituted by soft pitch. The proportions of binding medium were in both cases equal to 6 and 8 percent.

Figure 14 shows a graph, the curves of which are based upon shaped coke made from semicoke with caking coal as the only binding medium. The pressure and abrasion resistance of the coke are plotted in two curves, the course of which depends on the pressure applied while briquetting, as expressed by the vertical ordinates of the graph.

The mixture from which this coke was made consisted of 70 percent semicoke and 30 percent caking coal. The following conditions prevailed:

Semicoke: 15 percent volatile matter, 9 percent ash, grain size:
0 - 0.5 mm.

Caking coal: Grain size: 0 - 0.25 mm.

Raw briquettes: Moisture: 7 percent.

Temperature of briquetting mixture: 85° C.

Carbonization temperature of briquettes: 900° C.

From the graph of figure 14 can be deduced that the briquetting pressure applied is not of such decisive importance for the quality of the shaped coke as is generally assumed, while other factors characterizing the condition of the briquetting mixture have a considerably greater influence upon the properties of the shaped coke.

Results of Large-Scale Trial to Convert Coal from Upper Silesia into Shaped Metallurgical Coke

Table 3 contains all particulars of importance of the manufacture of shaped coke from coals of Upper Silesia produced by a trial carried out on a commercial scale. From the table can be deduced that the shaped coke represents, in chemical respects, a very valuable fuel that meets all demands that a metallurgical coke must answer. With contents of ash below 10 percent and an abrasion hardness exceeding 90 percent of this coke will satisfy all requirements, and it will be superior in use to the best coke-oven coke made.

Shaped Coke from Powdered Coal

To prove the independence of the two-stage carbonization process from the physical properties of the basic coal most plainly, powdered coal from Upper Silesia was used for manufacturing shaped coke by this process. The particulars of the operating conditions applied can be deduced from table 4, which indicates that the manufacture of a shaped metallurgical coke from such an inferior fuel was accomplished successfully. The abrasive resistance of the coke approached 84 percent very closely, and the results emphasize the fact that the two-stage carbonization process represents an ideal way to upgrade inferior fuels.

Comparison of Economy of Operation with Modern Coke Ovens

By a superficial comparison of the two-stage carbonization process with the one-stage operation of coke ovens, the first conclusion will be that, as regards capital charges as well as operating costs, the two-stage process will come out at a great disadvantage as compared with a coke-oven plant of equal capacity. In order to show that the differences are in reality much smaller than one is inclined to assume at the first moment, in the following a comparison will be drawn based upon conditions corresponding to actual operating practice based upon a throughput of 1,000 tons of coal per day equal to a coke production of 750 tons per day.

With chamber dimensions as applied to modern coke ovens, vis., length, 12 m.; height, 4.5 m.; and a medium width of 450 mm. and an average coking

time of 19 hours, 38 coke ovens will be necessary for the output stated. These coke ovens have a heated wall surface equal to $2 \times 54 = 108 \text{ m}^2$.

The chambers of the low-temperature carbonization ovens of the two-stage process have, with a depth of 6 m. and a height of 9 m., a heated wall surface of $2 \times 54 = 108 \text{ m}^2$ for each chamber. Compared with a throughput of 26 tons of coal per day of the coke ovens, having a coking time of 19 hours, a continuously operated, ceramic, low-temperature, carbonizing oven has a throughput of 60 to 68 tons, in average 64 tons of coal per day, so that, compared with the above 28 coke ovens required for the throughput of 1,000 tons of coal per day, only 16 low-temperature carbonizing ovens are necessary for the same capacity.

TABLE 3. - Shaped coke from coals of Upper Silesia

Origin of coals: I = Eminenz colliery II = Blechhammer colliery

1. Proximate analysis of semicoke

Constituent	Raw		Free of water		Free of water and ash	
	I	II	I	II	I	II
	Percent		Percent		Percent	
Moisture	0.9	5.3	0.0	0.0	0.0	0.0
Ash	9.6	7.7	9.7	8.1	0.0	0.0
Volatile matter	5.6	7.3	5.7	7.7	6.3	8.4
Fixed carbon	83.9	71.7	84.6	84.2	93.7	91.6
Total	100.0	100.0	100.0	100.0	100.0	100.0

2. Briquetting

Kind of coal:	I		II	
	mm.	Percent	mm.	Percent
Grain size of semicoke	0-1	59	0-1	67
Grain size of semicoke	1.0-2.0	41	1.0-2.0	33
<u>Mixture in relation to dry substance:</u>				
Semicoke	0-2	82	0-2	82
Coking coal	0-0.5	11	0-0.5	11
Pitch (softening point 83°C.)	0-0.25	7	0-0.25	7
Temperature of briquetting mixture...	$140-150^{\circ} \text{C.}$		$140-150^{\circ} \text{C.}$	
Pressure applied in atmospheres	200		200	
Pressure resistance of raw briquettes after 24 hours	19 kg/cm ²		54 kg/cm ²	

3. Carbonization

	I	II
Moisture in crude briquettes after 24 hours, percent	5.2	4.6
Temperature of carbonization, $^{\circ}\text{C.}$	900	900
Yield of shaped coke in relation to dry raw briquettes, percent	80.8	84.7
<u>Results of drum test (abrasion):</u>		
First run, percent	92	91.5
Second run, percent	90	92.6

TABLE 4. - Manufacture of shaped metallurgical coke from powdered Upper Silesian coal1. Low-temperature carbonization

Semicoke: Volatile matter: 4.01 percent Ash: 16.78 percent

Grain size of semicoke for briquetting		
Grain size, mm.	Single fractions, percent	Total, percent
0.00-0.25	21.8	21.8
0.25-0.50	14.0	35.8
0.50-1.00	18.3	54.1
1.00-2.00	22.2	76.3
2.00-3.00	11.5	87.8
3.00-4.00	12.2	100.0
Grain size of coking coal from Gleiwitz		
	100 percent	0-0.5 mm.
Grain size of pitch (softening point 76°C.)		
	100 percent	0-0.5 mm.

2. Briquetting and carbonizing

Temperature of briquetting mixture about 100° C.
 Pressure applied to briquetting mixture about 200 atmospheres
 Size of raw briquettes about 40 by 30 mm.
 Weight of raw briquettes about 35 grammes
 Volume of raw briquettes about 37 cm³ (cc)
 Corresponding to approximately per cm³ (cc) 0.95 grammes
 Charging temperature of raw briquettes about 900° C.
 Carbonizing temperature of briquettes 900° C.
 Coking time approximately 3 hours

3. Properties of briquettes made from different mixtures

Trial No.	Mixture consisting of -	Grain size, mm.	Pressure resistance of briquettes, atmospheres	Abrasion (drum) test	
				Above 25 mm., percent	Above 15 mm., percent
1	80 percent semicoke	0-4	Not determined	83.9	83.9
	10 percent coking coal	0-0.5			
	10 percent pitch	0-0.5			
2	90 percent semicoke	0-4	Not determined	51.1	51.1
	10 percent coking coal	0-0.5			
3	84 percent semicoke	0-4	17	71.7	71.7
	10 percent coking coal	0-0.5			
	6 percent pitch				

4. Properties of shaped coke from trial No. 1

Ash, percent 15.14
 Ignition point in oxygen, °C. 515
 Ignition point in air, °C. 565
 Apparent specific gravity, percent 0.915
 True specific gravity, percent 1.66
 Porosity, percent by volume 45

This great difference in capacity rests with the far-reaching utilization of the heat by means of heat exchanging and the transfer of the heat to the charge of coal. Such a method is not applicable in the coke oven, as the sensible heat of the coke as well as of the gases evolved from the coal must be removed without making use of it.

These, with respect to heat economy, very favorable operating conditions of the vertical, continuously operated, low-temperature carbonization oven, which naturally have a far-reaching effect upon the costs for putting down such plant, are also applicable for the high-temperature treatment plant that forms the second stage of the process. These chambers are also 9 m. high and 6 m. deep and are heated to a maximum temperature of 1,000° C. The throughput of such a chamber is equal to 42 tons of briquettes per day, so that for a production of 1,000 tons of coke per day 22 chambers are required.

The two-stage process thus requires, for a production of 1,000 tons of coke per day, 16 low-temperature carbonization and 22 high-temperature carbonization chambers of altogether 38 units all of equal dimensions, and the plant thus corresponds, in respect to the number of carbonization chambers, to a modern coke-oven battery of the same capacity. For the manufacture of a certain quantity of normal metallurgical coke from coking coal, the same number of coke ovens is required as is necessary to deal with these quantities by the two-stage process. The costs for the erection of these plants and the running capital charges do thus not differ considerably from each other.

In respect to the requirements of heat, it has been determined by calculation that 600 to 650 kcal per kilo (108-117 B.t.u./lb.) coal suffice to meet the heat consumption of the two-stage process. Compared with the 520 to 560 kcal per kilo (94-101 B.t.u./lb.) coal consumed in normal coking, the difference in consumption of heat is not large enough to play a decisive roll in respect to the economy of the manufacture of shaped coke. By trials carried through on an industrial scale, it has been ascertained that the low-temperature carbonization ovens of the first stage of the process had, in relation to their heated chamber walls, a capacity of 30 to 34 kilos coal per m² an hour. The heat consumption of the vertical, continuously operated, low-temperature carbonization ovens is, compared with coke ovens, considerably lower, for the reason that the amount of heat required for carbonization is much smaller. Compared with normal carbonization, coke and gas leave the continuously operated chambers having a temperature of 200° to 250° C. only so that the losses of heat are equal to about half of the normal heat of carbonization, amounting to 375 kcal (67 B.t.u./lb.). The heat requirements of 1 kilo of coal charged thus correspond, in a large industrial plant, to about 280 to 340 kcal (50-61 B.t.u./lb.) per kilo throughput.

In the second thermic stage of the process, the capacity of the heated chamber-wall surface is equal to 22 to 24 kilos coal per m² an hour and corresponds to about double the amount compared with coke ovens, and the losses of heat can be utilized by a correspondingly large volume of circulating gas, so that in the ovens of the second stage, also, the consumption of heat will not exceed 320 to 380 kcal (58-68 B.t.u./lb.) per kilo coal.

In table 5, the operating costs of a two-stage plant are set in comparison to a modern coke-oven plant of equal capacity, and for both the figures are based on a throughput of 3,000 tons of coal per day.

TABLE 5. - Comparison of operating costs of a two-stage plant and a coke-oven plant, respectively, both for a daily capacity of 3,000 tons coal

	Low temper- ature carboni- zation	Harden- ing	Produc- tion of shaped coke	Normal coke- oven plant
Throughput of coal per day tons	2,640	360	-	3,000
Semicoke per day do.	-	1,848	-	-
Coal-tar pitch per day do.	-	199	-	-
Abrasion of shaped coke, per day do.	-	233	-	-
Coke production per day tons	1848	2120	2120	2120
Production costs per ton of coke	RM	RM	RM	RM
Sulfuric acid, 27 RM per ton	-	0.07	0.07	0.34
Wash oil, 110 RM per ton	0.27	0.07	0.34	0.24
Electric power, 17.50 RM per 1,000 kw.hr. .	0.18	0.81	0.99	0.26
Water, 15 RM per 1,000 m ³	0.21	0.28	0.49	0.30
Compressed air	-	-	-	0.02
Steam, 2.50 RM per ton	0.15	0.38	0.53	0.63
Wages	0.44	0.71	1.15	1.04
Oven repairs	0.15	0.15	0.30	0.10
Upkeep	0.71	2.52	3.23	1.00
Rates and taxes	0.25	0.48	0.73	0.39
Capital charges	2.47	2.42	4.89	2.90
Production costs per ton of coke	4.83	7.89	12.72	6.64
To be credited per ton of coke:				
Small (abraded) coke	-	0.89	0.89	-
Coal-tar pitch, 41 RM per ton	2.04	2.07	4.11	-
Tar oil, 100 RM per ton	8.44	1.79	10.23	2.00
Ammonium sulfate, 80 RM per ton	-	0.18	0.18	1.11
Gasoline, 150 RM per ton	2.15	0.38	2.53	-
Benzol, 320 RM per ton	-	-	-	5.16
Gas, 18 and 17 RM per 1,000 m ³	1.25	1.56	2.81	3.81
To be credited per ton of coke	13.88	6.87	20.75	12.08

From the comparative figures compiled in table 5 it can be deduced that coke-production costs depend, first, on the price of the basic coal. The yields appearing in table 5, the manufacturing costs, and the amounts to be credited are based upon the manufacture of shaped coke from a geologically young coal with natural moisture and, on the other hand, on a coke-oven coke made from normal coking coal.

If the price for the coal to be treated first by low-temperature carboni- zation is to be equal to 11 M. per ton and the coking coal to cost 12 M. per ton, the total costs for the shaped coke amount to 16 M. and for the coke-oven coke to 13 M. per ton.

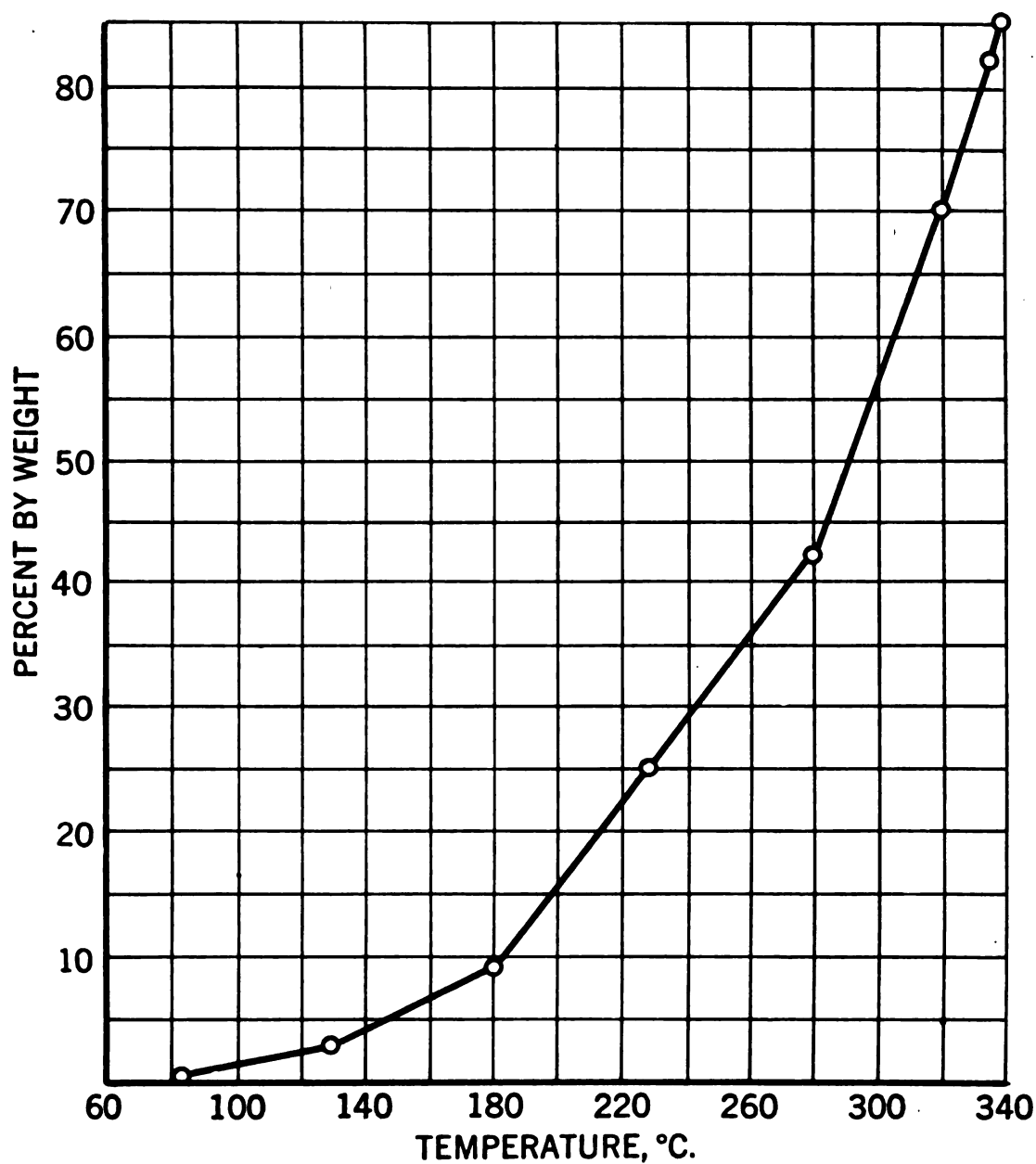


Figure 15. - Distillation curve of primary tar produced from coal in the continuously operated ceramic, vertical, low-temperature carbonization oven, by Didier Werke.

The higher value of the shaped coke, its even shape, dense structure, high reactivity, and its close, but for gases still easily penetratable, bulk, as well as the considerably reduced losses by abrasion justify the slightly higher production costs compared with those of the normal coke-oven coke. In many cases the higher production costs of the shaped coke will be more than compensated by the freights to be debited to the price of the coke-oven coke, which has to be transported over great distances, whereas the basic fuel enabling the production of shaped coke may be had within the vicinity of the consumer.

Direct Separation of Coal-Tar Pitch from the Gas

Contrary to coke-oven practice, the plants adapted for operating the two-stage process must be equipped with a briquetting plant as well as with a tar distillery in the latter case to recover the required pitch from the tar serving as a binding medium for the manufacture of briquettes.

The properties of the primary tar, as recovered from the continuous ovens of the first stage of the process, are compiled in table 6. The distillation curve indicating the course of the boiling points of the low-temperature tar is plotted in figure 15, from which can be deduced that the product represents a valuable tar, which, after the pitch has been removed, can be used as a fuel oil.

TABLE 6. - Boiling ranges of low-temperature coal tar

Fraction No.	Boiling range, °C.	Percent by weight	Specific gravity	Properties
Water		2.2	-	
1	85-130	2.4	Not determined	Stainless
2	130-180	6.2	Not determined	Light brown
3	180-230	16.6	0.963 (20°)	Do.
4	230-280	17.0	0.999 (20°)	Brown at 0° C.
5	280-320	27.6	1.002 (50°)	Partly solid
6	320-336	12.3	1.004 (50°)	Solid at 15° C.
7	336-340	2.7		Solid at 30° C.

	Percent by weight
	<u>84.8</u>
Residue and loss	<u>15.2</u>
	100.0

In order to effect this simple process of separating the oils and the pitch from each other without having to resort to a separate tar-distilling plant, which in respect to its initial as well as to its operating costs is comparatively expensive, it was remembered that many decades ago Walther Feld (34) had successfully effected a direct separation of oils and pitch as well as the extraction of tar from the hot crude-coke-oven gases. This was

accomplished by means of the centrifugal washer cleverly constructed and introduced by Walther Feld. It is intended to supplement the two-stage processing plant with a similar plant instead of a tar distillery. The great importance that must be attached to this kind of pitch recovery for the economical results of the two-stage process justified its brief description in the following by the aid of the diagrammatic flow sheet (fig. 16).

The crude gas coming directly from the low-temperature carbonization ovens of the first stage as well as from the high-temperature ovens of the second stage of the plant is drawn forward through the suction main 1 and into the box 2, serving to retain the condensed tar consisting principally of soft pitch leads the drain pipe 3. The gas main 1 is connected to the base of the centrifugal washer 4, the construction of which is so well known that it need not be further described in this place. The mechanical drive of the internal parts of the centrifugal washer 4 is effected by the electric motor 5 placed on the top of the washer housing and coupled direct to the vertical shaft of the washer by a special gear. The pitch separated out in the washer 4 passes through a syphon 6 into the pitch-collecting tank 7, to which is connected, on its base, the centrifugal pump 8 for the purpose of propelling the pitch still in its liquid state to the briquetting plant. The tank 7 is divided into two compartments, 7a and 7b, by means of a vertical plate, and the overflow pipe of box 2 leads into tank 7b, which at the same time serves as a circulating tank. Pump 9 is connected to the latter by which the three upper sections of washer 4 receive hot tar for washing the gas. The tanks 7a and 7b have steam coils in their bases, so as to be able to heat the contents.

The gas that has passed through washer 4 is subsequently treated in the same manner in passing through washer 10, but this unit can also take the place of washer 4 if the latter should temporarily drop out of operation by accident. To counteract the possibility of a drop of the gas temperature, a gas heater 11 is provided just before the inlet of the washer 10 which, by means of indirectly applied steam, raises the temperature of the gas again and can further serve as a temperature-adjusting arrangement. The drain pipe 12 of washer 10 leads to tank 13a, to which is connected pump 14, whereas the other compartment, 13b, of the tank is connected to the circulating pump 15, serving to feed the upper three sections of washer 10 with tar for scrubbing. The gas leaving washer 10 enters the water tube cooler 16, which consists of four gas compartments in series, each drain of which leads to a separate compartment of the collecting tank 17a - d.

The pump 18 drawing from compartment 17a transfers the oil to tank 13b or 7b, and it must be mentioned that tanks 7 and 13 are interconnected by an overflow, 19, into which a stop valve is interposed. The four compartments of water cooler 16 are, as already mentioned, connected to the four corresponding partitions, 17a - d, respectively, of collecting tank 17, each of which can be drained of water by a separate pipe connected to the common drain pipe, 20. The oils collected in tank compartments 17a - d can, by means of the pump 21, be transferred to storage tanks, or they may be used for flushing the collecting mains of the ovens by means of pump 22 so as to dissolve any deposits of soft pitch therein. Gas leaving cooler 16, having been freed of tar oils by condensing, now enters an oil scrubber to be stripped of light oils to be worked up into gasoline.



The gas is treated to remove separately the pitch and the tar fractions by adjusting certain temperatures and applying washing liquids having a certain condition, the circuit of which is closed by the mentioned connections of collecting tanks 7, 13, and 17.

The pump 9 forces high-boiling oil from tank 7b into the top section of washer 4, enabling distribution of this oil over several sections of the washer, at the same time. In the lower part of the washer the washing tar becomes enriched by the pitch constituents absorbed from the gas, and the pitch eventually collects in the base of tank 7a, from which it is drawn by pump 8 and transferred to the briquetting plant in the liquid state. Should the pitch not be hard enough and consist more or less of tar oils, it will, by stopping pump 8, overflow the partition wall from compartment 7a to 7b, and by means of pump 9 the tar will again be circulated over washer 4. The tar overflowing from the tank 2 is also conducted into tank 7b.

Although in washer 4 pitch only is being recovered, in washer 10, which serves at the same time as a standby unit for washer 4, a high-boiling tar oil fraction is recovered. Both washers 4 and 10 correspond exactly as to their construction, apart from gas heater 11, connected into the way of the gas just before it enters the washer 10. If used for the recovery of pitch, washer 10 is direct-circuited with collecting tank 13 and operated in exactly the same way, as described above by the aid of the pumps 14 and 15. In recovering heavy tar oil, a part flows according to requirements from the tank 13a into the tank 7b while the excess is transferred by means of the pump 14 to the tank for collecting heavy oil.

In the four compartments, a-d, of gas cooler 16 indirect cooling of the gas countercurrent to the flow of water through the cooler is effected, and in the four corresponding compartments, a-d, of collecting tank 13 the single fractions of the tar oils are collected separately.

The plant, started up for the first time, is adjusted so that the tar condensed in the gas main 1 runs by means of seal pipe 3 into the tank 2, from which it overflows into tank 7b. The tar is circulated through the washer until a certain equilibrium in the condition of pitch and oils is reached, recovering a pitch with the desired properties.

The plant represented by the flow sheet (fig. 16) serves to recover the tar in the form of pitch and several fractions of tar oils. If it is intended simply to separate pitch and tar oils as a whole and to dispose of the latter in the mixed state as fuel oil the plant can be constructed much simpler, as washer 10 will then not be required, and in a plain gas cooler the tar oils are subsequently recovered by condensing and are collected as a whole without separating into single fractions.

In the operation of this plant, the greatest importance must be attached to the condition of the tar circulated over the gas in the washer; also of great importance are the adjustment and maintenance of the temperatures of the gas and the oil, the best conditions, for which can be ascertained for each individual plant by practical trials only.

The direct recovery of pitch and tar oils represents a valuable supplementary arrangement for the two-stage carbonization process, because this additional plant supplies the necessary coal-tar pitch as a binding medium for briquetting the fuel without having to resort to the construction and operation of an expensive tar-distillation plant for this purpose.

SUMMARY

The attempts to produce a coherent hard coke from fuels independent of their chemical and physical properties, particularly from their agglutinating power, commenced many decades ago. The principle processes within the range of this development have been briefly outlined in the present treatise. While the manufacture of a hard coherent coke from lignite in one single stage depends on the application of an excessively high pressure in special briquetting presses developed for this purpose, different processes have been worked out for manufacturing a valuable shaped coke highly suitable for use as a metallurgical fuel. The effect of the chemical composition of the coke ash upon the operation of the blast furnace is taken into account. The manufacture of a metallurgical coke from noncoking coal had already been developed in the form of the American Carbocoal process. Though in itself sound, the process was a failure in technical and economical respects. For lignites as well as for noncoking coals, a two-stage process has been developed to manufacture a coherent and hard metallurgical coke of even shape. Both processes differ from each other by carbonizing the lignite in the first stage in unilow ovens at high temperatures, so that the hydrocarbons evolved are subjected to thermic decomposition. The true coal is in the continuously operated vertical ovens of the first stage treated by low-temperature carbonization, and in this stage thermic decompositions are prevented as far as possible to have the benefit of a high yield of primary tar. In treating the lignite, an aromatic tar is produced by carbonization in the first stage, whereas the tar recovered in the low-temperature carbonization ovens from true coal has an aliphatic nature. The two-stage process developed for the treatment of true coal is, in respect to its economical results, compared with a modern coke-oven plant of equal capacity. The constructional and operating details of such plants are described fully without it being possible to give an exact prescription meeting all circumstances encountered to produce a hard metallurgical coke from noncoking fuels. The properties of the coke are dependent upon so many interrelated factors that they can be determined and taken into account only for each individual case in order to produce a shaped metallurgical coke of the best quality that meets all demands made on such a fuel. To render such a two-stage carbonization plant independent from the erection and operation of a tar-distillation plant, the application of the Feld process for the direct recovery of pitch and tar oils separately from the crude gas has been taken seriously into consideration, and the description of such an installation concludes this treatise.

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APPENDIX CEXPERIENCES AND SUGGESTIONS AS TO THE ECONOMY OF RAW MATERIALS FOR THE
HYDROGENATION OR LOW-TEMPERATURE CARBONIZATION OF TRUE COAL AS WELL
AS FOR THE METALLURGICAL WORKING UP OF SMALL-GRAINED IRON ORESIntroduction

In the autumn of 1944 the following statement was published by the British government:

According to the statement made in the House of Commons by the Minister of Fuel and Energy, Mr. Lloyd George, the British government has caused thorough investigations to be made as to the technical and economical problems accruing in connection with the hydrogenation of coal. He added that during the war the coal tar produced at home had displaced fuel oil to a large extent in Great Britain. A further strong development of low-temperature carbonization is expected by the coal industry as a result of the experiences gained during the war and as an outcome of the present investigations, while the chances for the hydrogenation of coal are judged less positively. Even under considerably increased prices for petrol, the hydrogenation remains dependent upon financial support, while the British low-temperature carbonization industry already attained an economical footing and, on the strength of its byproducts, may form a new basis for promising industries. Apart from the development of the low-temperature carbonization of coal, the British government also mentions the enlargement of the British bases for the refining of mineral oils in order to prevent Great Britain from lagging behind in the field of the chemistry of oils and of the economic branches connected with it. The formation of a suitable research committee was already decided upon, and the members of this committee are at present elected.

This statement shows that the British government distinctly recognized the importance of the processes for upgrading coal and the scope of its future development and that the British government makes every effort to force an essential influence to expose in the following a few experiences gained in Germany in this field, and further to make a few suggestions which, mainly confined to the economy of raw materials, will be subdivided as follows:

1. Coal upgrading processes.
 - (a) Hydrogenation of true coal.
 - (b) Low-temperature carbonization of true coal.
2. The combination of low-temperature carbonization of true coal with the metallurgical working up of fine-grained iron ores, termed in this connection as "metallurgical carbonization."

- (a) Short delineation in respect to the economy of the raw materials.
 - (b) Advantages of the metallurgical carbonization process in respect to the economy of raw materials.
3. The coupling of hydrogenation and metallurgical carbonization.
 4. Prospects for the economic effects of the new processes and suggestions.
 5. Conclusive remarks.

Short Delineations of the Hydrogenation of True Coal in Respect to the Economy of Raw Materials

The balance of material of the hydrogenating process of true coal does not represent a favorable situation economically. The conditions which are expected of a coal to be hydrogenated are extremely severe and far-reaching and can be met only by heavy losses of raw material. Particularly the claim as to the necessary freedom from mineral matter, the contents of which are not to exceed 3 percent, compels the application of a coal-washing process causing a loss of "burning substance" in the intermediate products of the washery that will be equal to about 40 percent in relation to the raw coal. If it is further taken into account that this raw coal originates from the most valuable seams only, the disadvantage in respect to economy of the raw material is obvious. These far-reaching claims, which the operating practice demands of the composition of the coal to be hydrogenated, are one of the main reasons not only for the high production costs of the products of hydrogenation but are also, responsible for the fact that many coal-mining countries have so far refrained from installing coal-hydrogenating plants, as the coals available do not meet the demands made by this process as regards their quality.

The second very unfavorable cause, for reasons of economy of raw material, presents itself at the end of the hydrogenating process insofar as a very considerable portion of the coal substance treated by hydrogenation is discharged as so-called hydrogenation residue, and thus these tailings have partly escaped the hydrogenation. The proportion of this hydrogenating residue is largely dependent on the type of catalyser used and varies between 20 and 40 percent by weight in relation to the amount of coal treated. Based upon the raw coal introduced, only a comparatively very small proportion is actually converted into oil.

Apart from the excessive claims made with regard to the quality of the coal subjected to hydrogenation, further far-reaching demands are made to cover the consumption of hydrogen of the hydrogenating process. Although theoretically quite a number of water-gas processes are available for this purpose, the whole consumption of water gas is in practice covered by the gasification of high-temperature coke by the well-known method. In this connection must be taken into consideration that this coke could preferably be used for iron and steel works, and for its production the quality of the

coal must also satisfy certain claims. . . Since a large portion of the large-scale chemical processes are dependent upon the gas phase, and as a rule water gas forms its base, the demands on the coking industry from these consumers are quite important, and a considerable increase from this source may be expected. As the coking industry cannot be enlarged indefinitely, the use of coke for the production of water gas will partly impair the coke supplies of the iron and steel industries. Naturally, this can not be tolerated and forms, also, an obstacle against the further development of the hydrogenation of coal.

The very expensive apparatus and the very sensitive mode of operation of the coal-hydrogenating plants as well as their being easily subjected to accidents need not be mentioned here further as they effect principally the operating costs, whereas within the frame of this treatise principally the economic side of the raw material is to be taken into account.

On the whole, the experiences collected in Germany have proved that the hydrogenation of true coal on the lines carried out so far does not meet, by a long way, the moderate demands made in respect to the economy of raw material. The facts stated by the British government are thus fully confirmed by the experiences gained in Germany.

Brief Delineation as to the Economy of Material of the Low-Temperature Carbonization of True Coal

Contrary to the hydrogenation which aims at the complete conversion of the coal into oil, the low-temperature carbonization of the true coal simply effects a de-oiling of the coal to be treated. The yield of liquid products is thus exclusively dependent on the percentage of tar contained in the coal to be treated and, in the case of good bituminous coal, ranges between 10 and 12 percent by weight. Since in this treatment of the coal 70 to 80 percent by weight of the basic coal are, apart from the coal gas liberated, recovered as an oil-free solid residue, so, from a viewpoint of economy of raw material, this residual coke represents, without question, the principal product of the low-temperature carbonization process of true coal, whereas the primary tar can be taken as a byproduct only, though a very desirable one. The value of the semicoke as the residual product of low-temperature carbonization is the higher, the lumpier and harder the pieces are and the more their shape suits the purpose of its consumption, because this determines its value for use.

From these viewpoints result the following claims as to the quality of the coals suitable for the low-temperature carbonization process:

- (a) Economical. Contents of tar as high as possible, exceeding if possible 10 percent by weight.
- (b) Technical. Sufficient agglutinating power to form a lumpy residue, i.e., a semicoke of high value in its consumption.

Although the height of the contents of tar depends to the largest extent upon the nature of the coal and can only be taken into consideration by a

corresponding selection of coals, the newly developed processes of briquetting enable the manufacture of a shaped semicoke in a simple manner, and the shaping of the coke is far superior to the natural tendency of the coal to form lumpy pieces during carbonization. Thus, all countries, so far as they are in possession of deposits of bituminous coal, are now able to produce liquid fuels by means of low-temperature carbonization.

Experiences gained in Germany have further shown that true coal suitably briquetted and subjected to low-temperature carbonization will considerably increase the capacity of the carbonizing plant in comparison with its treatment of coal in a now-briquetted state.

Finally, extensive operating experiences have been collected that prove that the semicoke made from briquetted noncoking coals forms an excellent basic fuel for the manufacture of water gas, and can thus be used in unlimited quantities to fully satisfy the demands of fuels for producing water gas by the large-scale chemical manufacturers, so that they need not consume metallurgical coke on which the iron and steel industries have the first claim for its use.

As, contrary to high-temperature carbonization, the coal is subjected to distillation at a considerably lower temperature, the tar produced is almost free from compounds formed by thermal decomposition. This not only increases its yield considerably but effects a quite different composition, compared with coke-oven or gasworks tar. As in England, also in Germany it has been found that the low-temperature tar contains very interesting compounds suitable as raw materials for several new branches of chemical industry.

Compared with the plants for hydrogenation, the apparatus of low-temperature carbonization works is much simpler, and its construction does not call for special demands as to the quality of the materials. Great independence in the selection of the coals, as well as simple apparatus and mode of operation of the low-temperature carbonization process, assure favorable economic results.

Thus, also in regard to the low-temperature carbonization of true coals, the experiences gained in England principally correspond with the extensive operating experience gained in Germany.

Combination of Low-Temperature Carbonization of True Coal with Metallurgical Grading-up of Small-grained Iron Ores - Metallurgical Carbonization

Brief Delineation as to the Economy of Raw Materials

It is well known that the semicoke produced from true coals is, as regards its reactivity during combustion and gasification, greatly superior to high-temperature coke. For the expert it is quite obvious, and, by corresponding tests it has been amply proved that this superior reactivity corresponds to considerably increased reducing properties in connection with the smelting of ores. By utilizing this knowledge and applying the extensive experience that has been gained in producing semicoke from briquetted coal, metallurgical low-temperature carbonization has been developed which, in its

most pronounced form, represents a combination of (1) low-temperature carbonization of true coal (production of primary tar), (2) metallurgical smelting (production of pig-iron), and (3) manufacture of gas for synthesis (production of mineral oils).

Into the process for the low-temperature carbonization of coal applied in this connection, briquetting of raw material is incorporated. This indicates at the same time a very simple way to work up fine-grained ores, which, to enable their smelting in the blast-furnace, must be briquetted in any case. By adding the fine-grained ore to the coal just before briquetting the advantage of the increased reduction effect of the semicoke is gained in connection with the smelting process.

Briefly, the operation of the metallurgical carbonization is carried out in such a manner that fine-grained bituminous coal, which need not be a coking coal with agglutinating properties, is mixed with fine-grained or soft ores and briquetted. The briquettes are subjected to low-temperature carbonization. In the case of noncoking coals, a binding agent must be added, such as concentrated sulfide lye or waste hydrocarbons. The bitumen expelled from the coal and the binder during the low-temperature carbonization of the briquettes are recovered in the form of primary tar, which is worked up by distillation, yielding valuable mineral oils. As a residual product of the carbonization remains a briquette consisting of ore and semicoke free from tarry constituents. The ore is present in these briquettes in the magnetic state, clearly indicating that the originally present iron-oxygen compound has been largely reduced during carbonization.

These briquettes, consisting of a mixture of semicoke and fine-grained ore, would be readily suitable for charging into the blast furnace, but this kind of treatment would not take advantage of the reducing properties of the semicoke nor of the already effected partial reduction of the ore. In addition, the blast furnace would require further metallurgical coke, the consumption of which is to be avoided by the application of this process. In the blast-furnace process a lean, combustible gas is generated, which, on account of its composition and low calorific value, can not satisfy the large requirements of gas which the various processes of coal hydrogenation demand.

Considerably better results can be expected if these briquettes are subjected to complete gasification in a liquid slag producer of known construction or, better still, in a water-jacketed low-shaft furnace of special design. Gasification is accelerated by the even shape of the fuel. Under these circumstances, it is possible to produce, either in the liquid-slag producer or in the low-shaft furnace, a producer gas of continually even composition, which, as regards its calorific value, can be improved considerably if, instead of air for combustion, oxygen or air enriched with oxygen is introduced into the producer or furnace. Under these conditions, a gas free from nitrogen or with a low content of nitrogen but with a high content of carbon monoxide will be produced, which, by the well-known means of conversion and purification, can without difficulty be converted into a water gas suitable for synthetic purposes. By these means, new and extensive possibilities offer themselves to the large chemical industry by covering the huge demand of water gas for

synthesis, which so far could not always be satisfied completely for want of metallurgical coke.

During the complete gasification of the carbonized briquettes, their contents of ore are completely reduced and melted. This is effected without the aid of metallurgical coke, because, should the amount of carbon contained in the briquettes be insufficient, further amounts of lumpy semicoke may be introduced into the furnace or producer. In the same way, a further addition of lumpy ore may be effected if the excess of carbon in the charge is too big in relation to its contents of iron. Naturally, all other additions, such as limestone, etc., required to manufacture pig iron of the desired quality, can be made to the charge in the producer.

Advantages of the Metallurgical Carbonization as Regards Economy of Raw Material

Metallurgical carbonization, as already mentioned, is based upon the consumption of bituminous coals, fine-grained ores, and waste hydrocarbons, such as residuals from oil refineries, tar-distilling plants, hydrogenating plants, etc. The process offers new and very valuable possibilities, particularly for the utilization of hydrogenation residuals, which will be more fully treated later in this paper.

Because three vastly different processes are combined in metallurgical carbonization, such as low-temperature carbonization, metallurgical smelting, and the generating of water gas for synthesis, very extensive reductions of costs can be effected, not only in the construction of such plants but also in respect to the economy of raw materials.

The extent of advantages in the economy of materials gained by combining the mentioned production processes can be deduced from a comparison of the following figures.

Metallurgical Carbonization

The trial results available so far indicate that in smelting of fine-grained ores having an iron content of about 35 percent, such as the Minette ore from Lorraine, by charging 5 tons bituminous slack (Lorraine coal) and 1 ton waste hydrocarbons (hydrogenating residue), a yield can be expected equal to about 3.0 to 3.5 tons pig iron and about 1 ton primary tar, besides the synthetic liquid products without requiring the consumption of coking coal or of metallurgical coke.

Normal Requirements for the Same Production

Experience has shown that for the production of 1 ton of pig iron the following materials are required: 1.3 tons metallurgical coke or 1.8 tons coking coal. By carbonizing the latter, about 7.5 percent of tar is recovered.

Thus, for the production of 3 to 3.5 tons of pig iron, 5.5 tons coking coal are required, which on carbonization will yield about 225 kilos of coke-oven tar.

For manufacturing 1 ton of liquid products by chemical synthesis or by hydrogenation, 6 tons of coal are required, of which 50 percent must be coking coal. Thus, for manufacturing 750 kilos liquid products, 4 tons of coal are required, divided into 2 tons of coking coal and 2 tons of noncoking coal.

Comparison

To produce 3 to 3.5 tons pig iron and 1 ton liquid products, 9.5 tons of coal, including 5.5 tons coking coal, are required in the usual processes, and 5 tons of coal + 1 ton of waste hydrocarbons (not coking coal) are required by the new process.

The metallurgical carbonization of small-grained ores, by which process lumpy ores can be smelted at the same time, offers (compared with the blast-furnace process as the only means of producing pig iron from ores so far) numerous advantages, of which the most important are the following:

1. Departure from the exclusive consumption of metallurgical coke and, thus, independence from the supplies of coking coal and saving of the whole coke works. A corresponding saving of the losses of material in washing the coking coal (loss of middlings) and in the coking process the loss of ballast in breaking and classifying the coke (dust and sizes too small for use in the blast furnace.)
2. Extending the coal range for the manufacture of iron from the coking coal in the direction of the gas coal and similar sorts, which at the same time have considerably higher contents of tar than the coking coal.
3. Possibility of utilizing unwashed bituminous slack. By separating the smalls from the run-of-mine coal, cleaning of the remaining lump coal can be much improved as regards effect and costs.
4. Increase of tar yields in comparison to high-temperature carbonization, because coals with higher contents of tar in relation to coking coals are used, and by the low-temperature carbonization the yield of tar exceeds 90 percent as against 40 percent in high-temperature carbonization.
5. Simple working up and utilization of residuals from all refineries tar-distilling plants as well as the residuals from the hydrogenation of coal.
6. Reduction of capital outlay by the installation of liquid-slag gas producers or water-jacketed low-shaft furnaces instead of a blast furnace.
7. Reduction of costs for metallurgical working up by using cheap slack instead of metallurgical coke.
8. Improvement of the reducing effect by using an evenly shaped, highly reactive, semicoke of a size properly adapted to its purpose.

9. Generation of a producer gas with high calorific value by operating the furnace or producer shaft with oxygen. This gas can serve as a cheap basic fuel for the synthesis of motor spirit or for the manufacture of hydrogen for hydrogenating plants.

Coupling of Hydrogenation and Metallurgical Carbonization

Metallurgical carbonization represents a process that allows for the working up and profitable utilization of large amounts of hydrogenating residuals. This fact introduces quite new points of view in connection with the application of the hydrogenating process treating coal.

The mode of operation of the hydrogenation plants for true coal applied so far is exclusively directed to liquefy as large a proportion of the coal substance as possible at the highest efficiency, because so far everything discharged as residue must be with regard to the calculation of costs of the production of synthetic petrol it had to be eliminated among the items tending to reduce the costs. If a new and valuable way of utilization is opened, the so far minor and even troublesome waste product will become a desirable by-product, which correspondingly will be of interest as a factor assisting to reduce the costs of manufacture of synthetic petrol.

This leads to a further very interesting reciprocal effect in the economy of raw material. The adaptability of the hydrogenating residuals for the metallurgical working-up of ores is naturally the better the more carbonaceous substance they still contain, or in other words, the lower the hydrogenating efficiency of the plant from which they originate.

This claim corresponds completely with the actual run of the hydrogenating process. Like many chemical processes, the first 30 to 50 percent of the liquefaction of coal can, in the hydrogenation of fuel, be accomplished easily and with a small expenditure of technical means, whereas a further increase of efficiency in the rate of liquefaction can not be attained with a proportionate effort but at progressively heavy expense only.

Technically the highest efficiency and economically the best efficiency were opposed to each other so incompatibly, because a large loss of material in the shape of the tailings discharged from the hydrogenating process must be avoided. If by means of the metallurgical carbonization process a loss of material is prevented and, further, the production of a hydrogenation residual that has not been treated too far is even very desirable, it can now be justified to interrupt the hydrogenating process just at that point at which the run of the process would cause a strong rise of the progressive costs.

This train of thought, the practical realization of which can easily be possible leads to two important results:

1. The partial hydrogenation will in future, at least when coupled with the metallurgical carbonization process, take the place of the hitherto aspired complete hydrogenation of fuels.

2. A considerable reduction in the manufacturing costs of the total hydrogenation, as up to now these have justly been considered too high and incompatible with the economy of the process, raising at the same time the costs of the hydrogenating products to an excessive level. This reduction of costs will be accomplished by applying the partial hydrogenation, i.e., to terminate the process at a point where the progressive costs will make themselves felt, so that the heavily increasing effect of just this portion of the costs upon the height of the manufacturing expenses will be eliminated.

Supported by these facts, one is further justified to reduce in future the claims as to the quality, and particularly as regards a low content of ash in the coal which is to be hydrogenated, in a proportion that will have a far-reaching effect upon the economy of the material. Particularly the high and, in the long run, unbearable losses of material in the ashing of the coal to be hydrogenated, which so far seemed to be unavoidable, will be reduced considerably.

A further effect of this is that the number of kinds of fuel suitable for hydrogenation will be enlarged considerably. Also, the application of the most modern coal-cleaning process, as, for instance, the "thermal coal cleaning," by which, apart from the separation of mineral matters, a deduction of undesirable chemical constituents like oxygen, sulfur, etc., also is effected, will not be available and promises to become a further means for broadening the range of coals suitable for hydrogenation. Utilization of the remaining hydrogenation residue for the smelting of ores will be particularly important in those countries in which, in spite of large ore deposits, smelting of the ores could not be carried out for want of metallurgical coke. There is now a well-founded prospect of a satisfactory technical and economical success and even the utilization of large peat deposits by hydrogenation, and the production of pig iron can be contemplated.

Survey of the Present State of Iron Production of the World and of the Measures Taken So Far to Safeguard Its Future

Only those States were able to set up heavy industries and, on the basis of these, were able to develop themselves to be industrial countries that had within their boundaries not only rich and sufficient deposits of ores but also mines supplying the coking coals necessary for the manufacture of the metallurgical coke required to smelt these ores. The simultaneous presence of ore and coal deposits is rarely encountered, and the eminent economic power of the United States, England, Russia, and Germany rests with this value of rareness, as the deposits of only these countries contain both ores and coking coals. Larger geological deposits of coking coal are decidedly scarcer than deposits of rich iron ores, and this fact explains that, for instance, Belgium and Japan, which have no ore-mining industry worth speaking of, were able to develop an important iron industry, while, contrary to this, countries like Spain, Sweden, Brazil, Chili, Newfoundland, French North Africa, and others, in spite of their large iron ore deposits, could not develop a correspondingly important iron industry.

However, States possessing a large iron industry are not free from worries, because the future of iron production in none of these countries is assured indefinitely, but in several of these countries its end is foreshadowed. The cause of this rests with the fact that since in the first half of the 19th century the development of the production of iron assumed a tearing pace, so that the coking-coal deposits had to be worked in a robbing manner and at a corresponding rate, so that there is no country now with an abundance of coking coal, but in most of them a pronounced scarcity is acutely felt. (In connection with this must be taken into account that from 1800 up to the present day

the population of the earth has almost trebled, and during the same period the mining of coal has multiplied by 100 and the production of iron by even 200). Without coking coal there will be no metallurgical coke, and without this the operation of blast furnaces, the smelting of iron ores, and the manufacture of pig iron are impossible. The economical, political and military position of the leading industrial countries is endangered, and not only the responsible government quarters but also the heads of the mining and metal industries of the respective countries concerned are quite clear upon this point.

To safeguard the future production of iron, extensive research work was commenced about 20 years ago in all iron-producing countries, large sums of money being made available for this purpose. As a result, two problems have gained prominence: (1) Means to enlarge the basis of coking coals by a corresponding grading-up of the noncoking coals present in abundance, and (2) suitable pretreatment of the small-grained ores recovered in large proportion in order to save metallurgical coke for their smelting.

In the following, a short survey about the present state of the iron industry and the running research work in three leading industrial States, i.e., the United States, England, and Germany, will be given, while the situation prevailing in Russia cannot be taken into account for want of up-to-date and reliable information.

United States

About 85 percent of the iron ore mined in the United States originates in the deposits near the Great Lakes in the States of Minnesota, Michigan, and Wisconsin. Virtually only red iron ore is being mined. The quality of this iron ore is very uneven in physical and chemical respects and is characterized by frequent change of the layers, which already differ from each other by changing color. On the whole, the ores are soft and fine-grained and are justly termed "soft ore." Thus, the problem of dealing, with the fine-grained portion of the ore plays a prominent part in the metal industry of North America, particularly as every effort is made to introduce only an evenly sized burden into the blast furnace in order to maintain a high throughput. The largest sizes of the ore do not exceed 3 inches, and on the other hand as few fines as possible are desired. By means of breaking and classifying, the even sizes are obtained and at the same time a portion of the fine-grained ore separated by screening is converted by agglomeration into lumps of suitable size for charging into the blast furnace. Thus, during mining as well as during shipping, and eventually in breaking and classifying the ore, large amounts of fines cannot be avoided. The economical utilization of these, by saving metallurgical coke at the same time presents an important problem. Seen solely from the angle of utilizing the fines of the ores, the metallurgical carbonization process would represent a welcomed solution and is to fill a wide gap.

In addition, also, the present and particularly the future supplies of metallurgical coke required for smelting these ores does not appear to be very simple if, for instance, the remarks are taken as a basis which appear

in the report of Schmitz,^{5/} who during 1938 made an extensive trip through the mining and industrial regions of the United States by order of the mining industry of the Ruhr. The following sentences taken from this report are recapitulated:

For the future, the supply of suitable coking coals creates a real worry, and on this account already at the present time the range of good coking coals is extended by mixing different kinds of coal. The supply of coking coals, however, remains one of the most important technical problems, which can be solved only from the side of the materials. For this purpose, already systematic investigations, principally carbonization tests on a large scale, have been carried out, by which, also, petrographic knowledge is of particular account, without, however, having so far found a practical solution of the problem.

Not only in the United States but, as is well known, also in England and Germany intense petrographic investigations of coal have been carried out during the last 20 years. They have already thrown much interesting light on the single structural constituents of the coal, particularly in relation to their coking properties, but they have not been able to create a basis for the practical utilization of the results of these investigations on plants of a commercial scale. Without such a result, the requirements of practical operation are not met, as can be judged easily if taking into consideration that the coke-oven plant at Clairton, near Pittsburgh, consisting of 1,482 coke ovens has a daily throughput up to 3,200 tons of coking coal, and the coke-oven plant at the iron and steel works at Gary, near Chicago, consists of about 1,000 coke ovens, which must be charged regularly.

It is imagined that the process of petrographic separation as developed in Germany and applied to noncoking coals may be improved in time so that it will comply with the requirements of practical operation; it will not mean that it can fill its place on a very large scale to tackle a problem on the successful solution of which depends the future of the iron industry of all countries. But even if the solution of this problem as indicated should be accomplished, it would entail a considerable expenditure, and a future heavy increase in the price of the coking coals on account of the costs of the material and the economy connected with the petrographic separation could not be avoided. Every economist is acquainted with the fact that an increase in manufacturing costs of the iron-producing industries will in the long run affect all industries that depend on iron as their basic material.

England

England is virtually independent in respect to its requirements of iron ores, as there are not only important ore deposits at home, but England is

^{5/} Reisebericht über den Kohlen- und Erzbergbau der Vereinigten Staaten von Nord-Amerika mit besonderer Berücksichtigung der Rohstoffaufbereitung. Glückauf, 1938.

also able to choose among the different ore-exporting countries for the most favorable offers in technical and economical respects to cover satisfactorily the ore requirements of its iron industry. There exists in England, in spite of this situation, a problem as to the fine-grained ores, insofar as in the mining of the home ores, as well as during shipping and transportation of the imported ores, corresponding amounts of fines present themselves, which the English blast-furnace manager, just like his colleagues over the whole globe, are anxious to keep out of the blast-furnace burden. The usual means of agglomerating, sintering, or briquetting also are applied in England in order to be able to deal with the fine-grained ores in the blast furnace. It has already been pointed out that the application of the metallurgical carbonization process represents, compared with the processes so far practiced, a considerable technical progress.

The situation as to the supply of metallurgical coke for the iron industry in England is decidedly serious. Considering the small geological deposits of real good coking coal in England and the impossibility of satisfying the further requirements by buying on the world markets at any time, all carbonization problems have met in England for decades past with intense interest. In England the government has from the beginning taken a lead and provided special research institutes in which, however, the cooperation of the mining industry as well as of the universities and technical high schools plays a prominent part. Apart from the Fuel Research Board's special department for carbonization, three special societies to study questions of carbonization were founded years ago and are financially well supported; i.e., one for Yorkshire and the Midlands, one in the North of England for Durham and Northumberland, and one for Scotland. Also, the institutes for fuel technology affiliated with the technical high schools are handling the same problems. A net of co-operators and experts divided over the whole industrial area is taking care that the research work always keeps in close contact with practical operation to meet its direct wants.

Since the coal seams in England consist chiefly of coals containing 30 to 35 percent volatile matter, the degree of coalification does not indicate their coking properties, as with the coals of the Ruhr district, and for this reason the petrographic study of the coals is taking a prominent part in the research work with the expectation that in this way progress will be achieved. Thus, in England, as in North America, the solution of a decidedly important problem is sought in principally the same direction for assuring the further production of metallurgical coke; and, as already mentioned, extensive work of the Ruhr coal industry and the coal-research institutes supported by it has been carried on for many years past with the same object in view. But in England, also, no solution has yet been discovered which would enable the conversion of the noncoking coals by means of petrographic separation on a large commercial scale in such a way that they would be suitable to be charged into the coke ovens to procure a good metallurgical coke.

Germany

Similarly, like in England, the manufacture of pig iron in Germany also depended on the extensive use of foreign ores with high contents of iron, particularly Swedish ores, though there are extensive iron-ore deposits in Germany which, however, have comparatively low contents of iron. Especially the Lorraine Minette repeatedly played a prominent part in the German iron-ore supply, as up to 1918 about four-fifths of the German iron ore mined consisted of Minette, and since 1940 Minette has again formed the principal portion of the German iron-ore output.

During 1935, autocratic efforts and valuta difficulties were the reason for manufacturing pig iron, depending to a large degree upon German ores, and for this purpose the iron-ore deposits situated on the north boundary of the Hartz Mountains were developed in order to meet the demand of the newly constructed large works at Salzgitter. By applying there at the same time a new German process which had already before been tried in England, it is possible to obtain a good pig iron from these ores with high contents of silicon. Also, the rest of the German metal industry has, for different reasons, followed in the same footsteps and melted, to an increased extent, German iron ores of this kind in order to become independent from the import of foreign iron ores, although they offer the advantage of having higher contents of iron.

Naturally, the smelting of such ores with low contents of iron and containing, in addition, mineral matter which, as ballast, tends to upset the normal operation of the blast furnace, causes a considerable rise of costs compared with the smelting of the good-quality ores rich in iron used so far. This undesirable rise of costs, which in the end will affect the whole structure of prices of the industry, depending on iron as a basic metal, induced the iron industry to the effort to counteract the rise of costs from the blast-furnace and operating end. In the construction of standardized blast furnaces with a daily output up to 1,000 tons of pig iron and smelting ores rich in iron, as well as by contemplating the construction of large blast furnaces with a daily output up to 2,000 tons of pig iron, hopes are entertained that this is the right way to reduce operating costs. There are a number of questions in connection with the construction of new blast furnaces that cannot be answered clearly, as, for instance:

1. Is the shaft of the large spaced blast furnace to be made of circular section, or, to prevent the formation of dead space, is the section to be of oval shape?
2. In case of an oval section, what means are applied to distribute the burden evenly over the large sectional area?
3. By what means are the large quantities of burden and coke to be conveyed to the furnace mouth?

Apart from these questions, the main problem is not solved as to how the increasing hardness of ore and coke, which must be raised together with the rate of throughput, is to be achieved by the ore mines and the coke-oven

plants. As long, however, as such vital questions cannot be answered conclusively, the economic effect of smelting German ores in increased quantities, which in itself may be unavoidable, cannot be considered to be satisfactorily cleared up.

In addition, further considerable difficulties are encountered, and these are, on the whole, the same as those which have already been mentioned in connection with the carbonization and smelting processes of the United States and England. Also, the German mining and metal industry has every reason to study carefully the problems of the metallurgical carbonization process and the broadening of the range of coking coals in order to safeguard the future of pig-iron manufacture. This, in fact, has been done for years now on a broad basis, but without noticeable decisive results.

In mining the German iron ores at home, particularly those at Salzgitter with low contents of iron, the problem of the fine-grained ore can be said to be of special importance, because the larger part of these ores is soft and powdery and has a strong tendency to disintegrate. Since, in comparison to the foreign ores rich in iron and easily smelted, the poor German ores, even consisting of lumps, demand in any case a considerably higher consumption of coke in the blast furnace, the efforts should be doubled to smelt at least the fine-grained ores without the use of metallurgical coke.

Extensive trials with the metallurgical coking process have proved the German ores with low contents of iron can, in spite of their relatively high contents of silicon, be smelted without the use of metallurgical coke and at a high rate of throughput. These trials proved, further, that in the course of the new process it is quite possible to treat large quantities of lumpy ore at the same time in the low shaft furnace together with briquetted fine-grained ore. In the example stated above under the heading Effects of the Political Economy upon the New Processes, Suggestions, and Their Prospects, this possibility has been made use of. The trials carried out since in this direction were so successful and convincing that the construction of a large trial plant to be erected at the Salzgitter works, having a daily throughput of several hundred tons, had been contemplated, and it was further planned to apply the metallurgical carbonization process utilizing noncoking coal instead of enlarging the blast-furnace plant by an additional 16 units, according to the original intention. In any case, based upon the results obtained, the metallurgical carbonization process will be suitable and destined to offer considerable support to German ore mining and to the use of German iron ores.

As already mentioned, extensive research work is also conducted by the German coal-mining industry, in which the Ruhr district is leading. This research proceeds in the same direction and partly, also, in the form of an exchange of ideas and experience with mining and other research institutions of the United States and of England. Although the Ruhr area contains the largest European coal deposit, it can, as a result of the extensive mining that has gone on for several decades now, not be maintained that there exists, even in the Ruhr district, an excess of coking coal, while, as is well known, the coals mined in the Saare region, as well as the Upper Silesian deposits, possess none or very small properties required for the manufacture of metallurgical coke. The efforts to broaden the range of coking coal and to find a way

to convert the noncoking coals abundantly present in all coal districts, particularly the gas coals, into a good metallurgical coke by means of suitable pretreatment, will readily be understood and must also in future be supported by all means, though the result of the research work carried through in this direction has not yielded a solution of the problem that can be applied in practice and will meet its demands.

SUMMARY

Coal mining and the manufacture of iron are the basis of the whole industrial life, and the commercial, political, and military power of the industrial States leading at present is based upon circumstances provided by nature, according to which coking coals and iron ore must be abundantly at disposal within their own boundaries and be thus within their power.

Since at the end of the 18th century the vanishing charcoal could, for smelting ores, be replaced by metallurgical coke manufactured from true coal, no change of any consequence can be noted in connection with the underlying processes for smelting the ores, in spite of the many individual technical progresses. Also, at present, just like many decades ago the normal operation of the blast furnace depends principally upon the size of the ore and of the coke and depends thus upon their physical condition.

In all iron-producing countries, the deposits of well carbonizing coals have since diminished to such an extent that in the near future the production of a solid and hard metallurgical coke is questionable. In the search of means to artificially broaden the range of coking coal, extensive research work was started years ago, but this was based principally on the knowledge of the petrography of coal and aims at the development of a process for the separation of noncoking coals into their individual components, at the same time discharging those component parts that are detrimental to the agglutinating power. This research work has, without doubt, been leading to very valuable knowledge in connection with the petrography of coal, and very interesting trial results on a laboratory scale also have been observed. The results so far obtained, however, are insufficient to satisfy the requirements of a plant operating on a large commercial scale, and there is no prospect in sight if and when a change in this direction may be expected.

On the other hand, it must be kept in mind that the solution of the cited problem gradually becomes an urgent necessity, because the geological deposits of coking coal diminish from day to day, and in some countries the time can already be fixed when serious difficulties in the provision of fuel will have to be faced by the operation of blast furnaces.

Thus, the question becomes urgent, whether, in respect to the importance of a secured manufacture of pig iron, it is not advisable, without impairing the full speed of progress of the research work commenced since for developing the petrographic separation of noncoking coals and thus for maintaining, also, in the future the coke-oven and blast furnace operation as practiced so far, to apply means that differ entirely from the present ones and, by introducing, for instance, the metallurgical carbonization process, to become quite

independent of the consumption of coking coals and at the same time of the whole coke-oven and blast-furnace operation as practiced so far. This means a departure from the whole process in the present manufacture of pig iron and to carry out the future smelting of ores in the low shaft furnace by consuming noncoking coals as a fuel. All difficulties in the provision of raw materials experienced so far which oppose the maintenance of the present manufacturing process for securing the future production of pig iron and which, without question, form a very serious danger for the whole economical position of the present iron producing countries would at once disappear by the introduction of the metallurgical carbonizing process.

The results of such a change in the manufacture of pig iron would have an effect almost impossible to predict. So, for instance, would, apart from the blast furnaces, also most of the coking plants installed at iron works disappear, among these, also, the above-mentioned coke-oven plants at Clairton and Gary in the United States, which can be considered the largest in the world. They would then have been overcome by more modern processes. The moment of inertia which is particularly effective in the production of bulk materials will, in the construction of the required new plants, prevent also in this instance the enormous capitals invested in the present production plants from suffering by impairment of their intrinsic value as well as their interest. Also, an introduction of the metallurgical carbonization process step by step would contribute to effect, within a short time noticeable relief of the existing difficulties in securing the necessary raw materials for the running manufacture of pig iron. Since, on the other hand, noncoking coals and iron ores of minor value as to their physical condition are amply at disposal in many countries, the metallurgical carbonization process could pave the way for a considerable geographic extension of the manufacture of pig iron.

During the last decades liquid fuel has appeared as an even partner to coking coal and iron, which so far formed the only pillars of industrial economic life, because no country can hold its position, with the hope of success if it should suffer by a deficiency of liquid motor fuel. It has been shown that the metallurgical carbonization process can also, in this respect, be applied with great advantage, quite irrespective of whether the recovery of primary tar or the manufacture of liquid fuels by means of the synthesis of water gas, or eventually the hydrogenation as practiced so far, for the partial hydrogenation will, apart from the important advantages in the respect to the economy or raw materials, cause a considerable reduction in the costs evolved by the total hydrogenation. They have, as mentioned in the beginning of this treatise, been declared as excessive and too high for liquid hydrogenation products by the cited statement of the British government. By the manufacture of liquid motor fuels, together with a valuable carbonaceous material for the reduction of iron ore in the shape of hydrogenation residuals, the hydrogenation process extends its scope to a far-reaching problem and secures its important position within the range of the various processes for the upgrading of coals.

Since coal, iron, and liquid fuels are, apart from the nutrition, the principal and material basis of every State economy, the new process and suggestions expounded above could, suitably applied, produce a considerable economical and political effect not only at home but also internationally.

PART II. - SLANTING-TYPE DIDIER COKE OVENS, STADTISCHE WERKE KARLSRUHE^{6/}

INTRODUCTION

The information given below was obtained in answer to questions submitted both in questionnaire form and verbally. Unfortunately, tracings and blue prints of the details of construction of these ovens had been removed or destroyed by the invading armies and were not available. This investigator endeavored to obtain details of design of these ovens from Dr. Adolph Thau of the Didier Werke, Berlin, but his files, also, had been completely evacuated by the Russians. However, upon request, the sketches of the oven chamber included at the end of this report were prepared by the plant personnel under the direction of the works manager. Although these sketches are not as complete as would be desirable, they do show the principal features of the oven design.

Gas, coke, tar, and benzene are manufactured at this plant from coals obtained from the Saar, Ruhr, and Eschweiler districts. War conditions had forced the management to use only coals from the Saar, with the result that a poor quality of spongy, soft coke was obtained.

Of particular interest here is the use of the sloping type of Didier gravity discharging coke ovens. Plant now running at 28 percent of present capacity. Thirty-five of the seventy coke ovens are in condition to operate. Plant was damaged badly by bombs; the Gas Institute Laboratory and equipment in it were completely destroyed.

COKE OVENS

Between 1927 and 1936, three batteries of sloping-type, Didier, gravity-discharging, coke ovens were built. The batteries contained 30, 15, and 25 ovens, respectively. Thirty-five of the seventy ovens are in condition to operate. Ten of these are operating on a 48-hour cycle, and ten more are due to go into production in the near future. Nine were using coal, and one was using wood on account of the lack of availability of coal in September 1945, and these were running on a 48-hour cycle.

These ovens have two distinct advantages. They can be built where space is not available for the more common slot-type oven, on one side of which space must be provided for a full-length pushing ram. Also, as the coke is discharged principally by gravity, one may carbonize poorly coking coals and discharge the weak coke which could not be pushed from a slot-type oven without crumbling and ruining the side walls of the oven chamber. Other installations of this type of coke oven are operated at Mainz, München, Halberstadt, and Immendingen.

Although the platform for the pushing ram and the door into the oven through which it enters are indicated in the accompanying sketches, the

^{6/} Prepared from Joint Intelligence Objectives Agency report "Slanting Type Didier Coke Ovens, Stadtische Werke Karlsruhe," by Frank H. Reed, 1945. (FIAT Final Report No. 566.)

pushing ram for the ovens is not shown. It is used only to loosen the charge and start it on the way out of the oven, as it will go the rest of the way by gravity. Total length of the ram is about 5 meters, and it is jointed at about 1-meter intervals. It is fitted into a vertical guide, the bottom end of which is bent at an angle of 37° to permit the ram to enter parallel to the oven floor.

In the accompanying sketches there is indicated, also, a steam inlet for the production of blue water gas. However, these ovens are not used for the production of water gas. Separate units in another part of the plant are used for this purpose. Although these ovens were designed originally to produce blue water gas from the coke at the end of the coking period without further handling, it was found that the shrinking of the charge in the oven caused the steam to channel and bypass the coke, and that the design was not satisfactory for the production of blue water gas.

Each chamber is 7,600 mm. long, 3,500 mm. deep, and 430 mm. wide. Chamotte (ceramic) and silica brick are used in oven construction. Each five chambers is heated from one oven. Seventeen percent of the coke produced is used in underfiring. Normal consumption is 250 tons coal per day, producing 200 tons coke. Present consumption is only 60 tons coal per day.

COALS USED

Before transportation facilities were disrupted, the Company used gas coal from the Saar and coking coal from the Ruhr and Eschweiler districts. A normal blend contained 60 percent Saar and 40 percent Ruhr coal. In September 1945 only Saar coal was used. Typical analysis of the coals used (identified by area and mine) are given in table 1. The bulk density of the Ruhr and Eschweiler coals is 0.790 tons per cubic meter while that of the Saar coals is only 0.736 tons per cubic meter.

OPERATION

From each kind of coal delivered to the gas works an average sample is taken and analyzed for moisture ash and volatile matter. The coal is either stored or moved directly over a coal breaker and mixed into the gas producing chamber. An average sample is taken again after the mixer and analyzed for moisture content.

The temperature of the ovens and chambers are checked daily by means of an optical pyrometer. The combustion gases pass over the heating elements of the boilers before entering the chimney. In doing so the gases are cooled from 450° C. to 220° C. The resulting coke is quenched with water and conveyed mechanically via the coke breaker and sizing apparatus into the storage bunkers. Average samples of the coke produced are taken daily from each size of coke. These samples are analyzed two or three times per week for ash, moisture, volatile matter and calorific value.

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TABLE 1. - Coal sources and analyses (average analyses 1943)

Mine	Size of coal	Percent								
		Crude moisture	Hydrosc water	Total water	Raw coke	Ash	Pure coke	Volatile	Pure coal	Ash in the raw coke
<u>Ruhr District</u>										
Rheinbaden	Fine	9.8	0.4	10.2	71.3	7.3	64.0	18.5	82.5	10.2
Rheinpreussen	Lump	2.7	0.6	3.3	69.1	4.9	64.2	27.6	91.8	7.1
Rheinland	Gas (unclassified)	3.4	0.7	4.1	65.8	8.9	56.9	30.1	87.0	13.5
Shamrock 3-4	Fat (unclassified)	2.3	0.4	2.7	78.2	11.8	66.4	19.1	85.5	15.0
Fr. Heinrich	Nut I	1.7	0.4	2.1	74.9	7.0	67.9	23.0	91.0	9.3
Harpen	Nut II	2.5	0.3	2.8	72.6	3.5	69.1	24.6	93.7	4.8
Hugo	Nut III	3.2	0.2	3.4	75.7	6.8	68.9	20.9	89.8	9.0
Victor	Fat nut IV	1.7	0.4	2.1	74.9	7.0	67.9	23.0	91.0	9.3
<u>Saar District</u>										
Maybach	Fine	11.5	0.5	12.0	58.8	11.1	47.7	29.2	76.9	18.9
Heinitz	Rough	2.5	0.6	3.1	63.0	5.2	57.8	33.9	91.7	8.3
Jagersfreude	Breeze	3.3	0.5	3.8	68.3	16.6	51.7	27.9	79.6	24.1
König	Lump	2.1	0.4	2.5	64.1	5.2	58.9	33.4	92.3	8.0
Kleinrosseln	Nut III	2.0	0.6	2.6	62.8	5.7	57.1	33.4	91.7	9.1
<u>Eschweiler District</u>										
Anna	Fine	9.9	0.4	10.3	74.1	8.4	65.7	15.7	81.3	11.3
Anna	Nut IV	3.5	0.3	3.8	79.2	7.1	72.1	17.0	89.1	9.0
<u>Upper Silesian District</u>										
	Coke fine	3.5	1.0	4.5	62.2	6.1	56.1	33.3	89.4	9.8

The raw gas which is produced is sucked out of the coke oven batteries, cooled in the Reuter cooler (water cooling) and separated from tar by shock condensation (Pelouze). The ammonia is washed out in rotating standard scrubbers. The naphthalene contained in the raw gas is picked up by the benzene washing oil while the benzene is being washed out. The hydrogen sulphide contained in the raw gas is removed by a dry cleaning process by natural or synthetic iron ores (Lux or Lauta mass). The gas is measured by a rotating gas meter before storage.

COKE

Under normal operation (1943) approximately 4.8 tons of coke were received from six tons of coal. The coke sold was used for the following purposes:

<u>Use</u>	<u>Percent</u>	<u>Size</u>
For metallurgical purposes	2	Over 90 mm.
For central heating	60	40-90 mm.
For heating stoves	30	10-40 mm.
For brick ovens	8	0-10 mm.

Ash, moisture, calorific value and melting point of the ash of the coke were determined. Also shatter and tumbler tests were run. For the volatile matter determination the coke was ground to pass through a sieve having 900 meshes per square centimeter and then heated at 850° C. The tumbler test is made in a steel drum 1,000 mm. in diameter and 1,000 mm. long into which are welded four angle irons each 90° apart. The drum rotates 100 revolutions in four minutes. For each tumbler test are used five kilos of coke sized between 40 and 60 mm. This coke has passed the shatter test before being used in the tumbler test. Other tests which are made occasionally include porosity, ignition temperature, reaction ability and reduction ability. These latter tests are of slight interest at the Karlsruhe plant as most of the coke is used for household purposes.

In September 1945 the plant used only coal from the Saar, operated under a coking cycle of 48 hours instead of 24 hours; operated at a temperature of 900° C. instead of 1,200° C. and produced a soft spongy coke of inferior quality. Formerly, under normal operation the drum stability factor was 70. In September it was 55 to 56. Formerly, the breeze amounted to 5 to 8 percent. In September 15 to 18 percent was received. The breeze is that coke passing through a 7 mm. screen. Whereas, formerly, 20 percent of the coke went to industry and 80 percent to household uses, now only 3 percent goes to industry.

As of the date of this report the plant is back on a normal operating cycle of 24 hours and is using coal from the Ruhr.

BYPRODUCTS

Gas

Under normal operation (1943) the yield of gas was 410 cubic meters per ton of coal. At present (1945) some wood is used and the gas from it is not separated from that produced from Saar coal. In table 2 is recorded the

results of gas analyses obtained in April 1943 (normal operation) and in August 1945 (present operation). The chief differences are: Present operation gives a gas with (1) a lower hydrogen content; (2) a higher nitrogen and carbon dioxide content, and lower calorific value.

TABLE 2. - Gas analyses

Date:	April 1943				August 1945			
	5th	15th	25th	Average	6th	13th	29th	Average
Gas, percent:								
CO ₂	2.6	3.0	3.2	2.9	6.9	5.0	6.4	6.1
C _m H _n	2.4	2.5	2.4	2.4	1.2	1.0	1.0	1.1
O ₂	0.3	0.5	0.4	0.4	0.8	0.7	0.8	0.7
CO	5.4	7.4	6.0	6.3	5.9	9.0	9.4	8.1
H ₂	47.5	42.6	48.0	46.0	33.4	35.3	34.2	34.3
CH ₄	27.4	27.8	25.6	27.3	25.0	24.0	24.4	24.5
N ₂	14.4	16.2	13.4	14.7	26.8	25.0	23.8	25.2
Calorific value:	4,350 kg cal/N m ³				3,900 kg cal/N m ³			

Oil

9.4 kilos of crude benzene were produced per ton of coal. From this crude benzene was produced fuel for internal combustion engines, purified toluene and solvent benzene (boiling point 150° C., boiling limit 200° C.). The toluene was sold to the producers of explosives and the solvent benzene to paint manufacturers.

Tar

Each ton of coal yielded 35 kilos of tar of specific gravity 1.15 and a moisture content of 4 percent. The viscosity of the tar (Mallison at 40° C.) was 34.7. The tar was sold to tar distilleries as there was no tar distilling equipment at Karlsruhe.

The tar analyzed as follows:

Material	Boiling range	Percent
Light oil	Below 170° C.	0.24
Medium oil	170-230° C.	3.30
Heavy oil	230-270° C.	12.21
Anthracene oil	270-350° C.	21.73
Total oil	-	37.48
Pitch residue	-	62.52
Phenol content of the tar ..		3.5

Capacities of units of equipment:

Number	Type of units	Capacity in cubic meters, m ³	Time, hours
2	Water coolers, capacity of each	50,000	24
1	Water cooler	100,000	24
2	Gas suction pumps, capacity of each	50,000	24
1	Gas suction pump	100,000	24
2	Tar separators, capacity of each	50,000	24
2	Ammonia scrubbers, capacity of each	150,000	24
2	Benzene scrubbers, capacity of each	70,000	24
3	Cleaning chambers for hydrogen sulphide, total capacity	150,000	24
1	Rotating gas meter	150,000	24
1	Wet-station gas meter	50,000	24
3	Gas holders (content)	40,000	
		28,000	
		20,000	
2	Pressure regulators, capacity of each	75,000	24
2	Gas compressors for medium pressure (2,500 WC=0.25 atm.), capacity of each	80,000	24
2	High-pressure compressors, 2 atm., for dis- tance pipeline gas, capacity of each	75,000	

COST FIGURES AND SALES PRICES

The following cost figures and sales prices only were available at the time of the trip:

Production cost of gas	3.8 Rpf/m ³
Basic price of coal	RM 22.50/ton
Average sales price of coke	RM 24.-/ton
Sales price of tar	RM 44.-/ton
Sales price of benzene	RM 420.-/ton
Average sales price of gas	RM 0.11/m ³
Spent sulphur cleaning mass	RM 23.-/ton

LABOR

The following figures on labor requirements, in man hours, do not include labor for auxiliary departments such as power house, repair shops, garages, and general labor:

Coal transport and preparation (capacity 40 tons of coal per hour, requiring 7 men to handle)	0.175 man hours per ton of coal.
Production of coke	0.25 man hours per ton.
Gas cooling and cleaning	0.3 man hours per 1,000 cubic meters of gas produced.
Benzene production	1.1 man hours per 100 kilos of raw benzene.
Gas compression and storage	0.3 man hours per 1,000 cubic meters of gas produced.

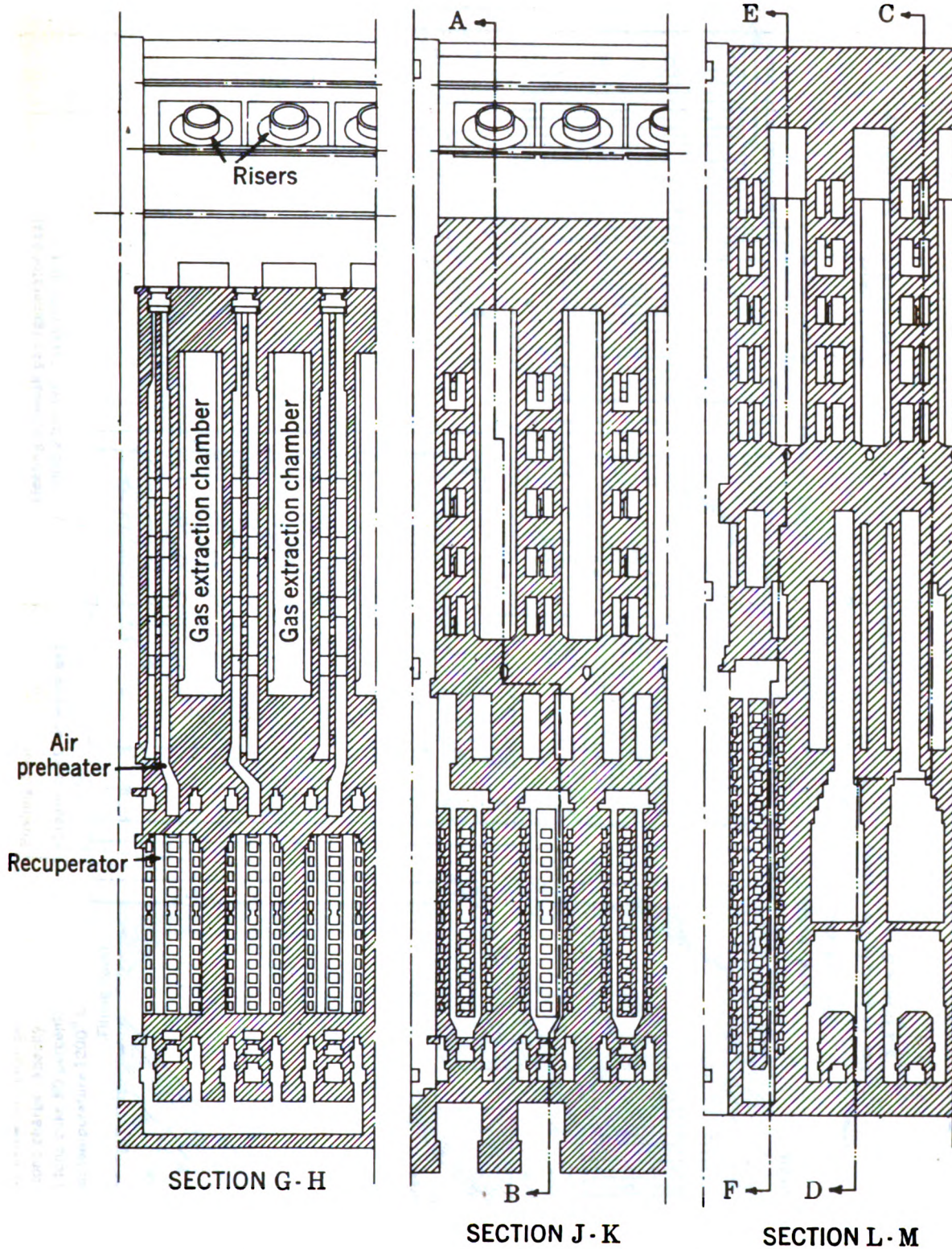


Figure 17. - Cross sections of the inclined chamber-oven construction, Didier gas plant, Karlsruhe.

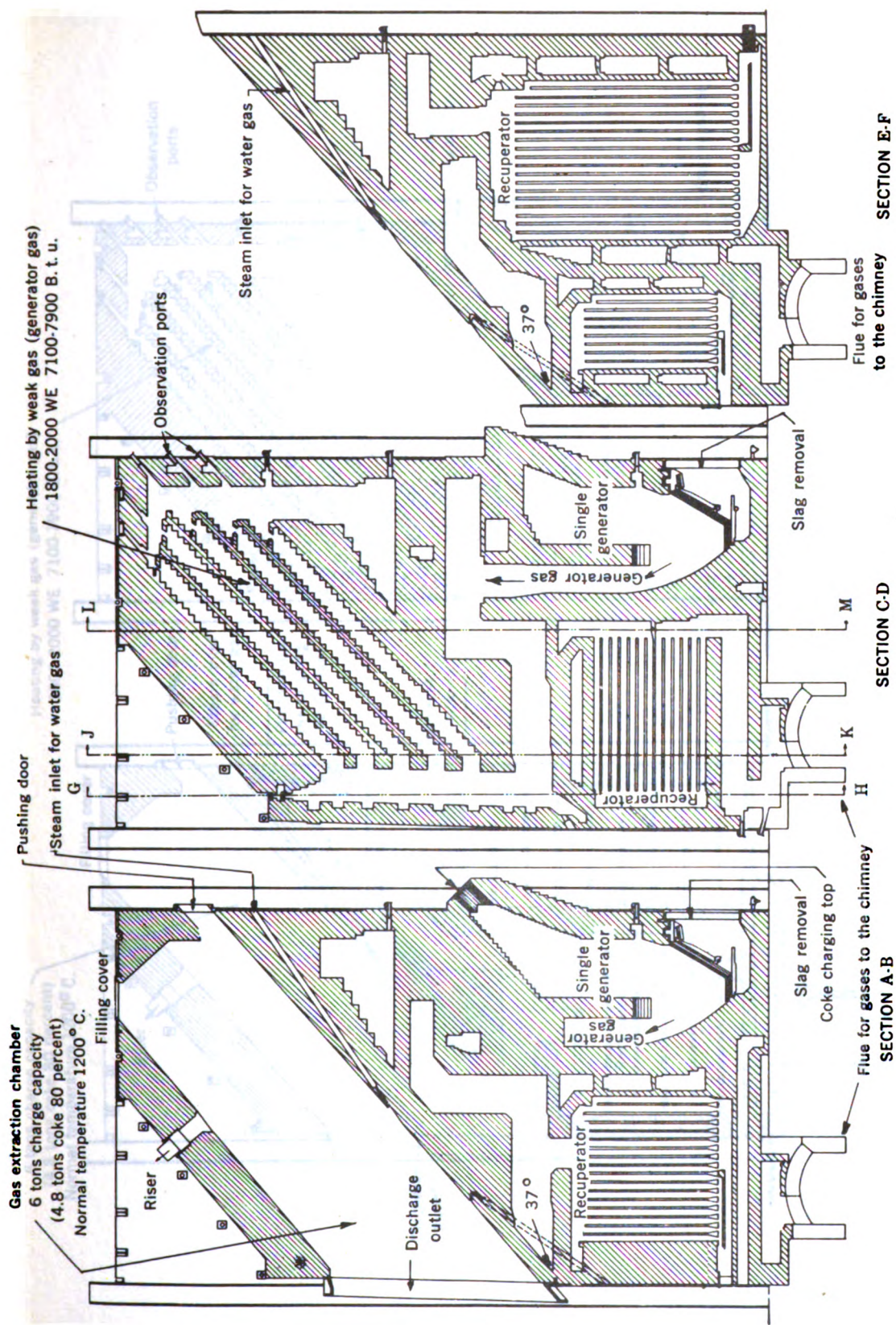


Figure 18. - Inclined chamber oven construction. Didier gas plant, Karlsruhe.

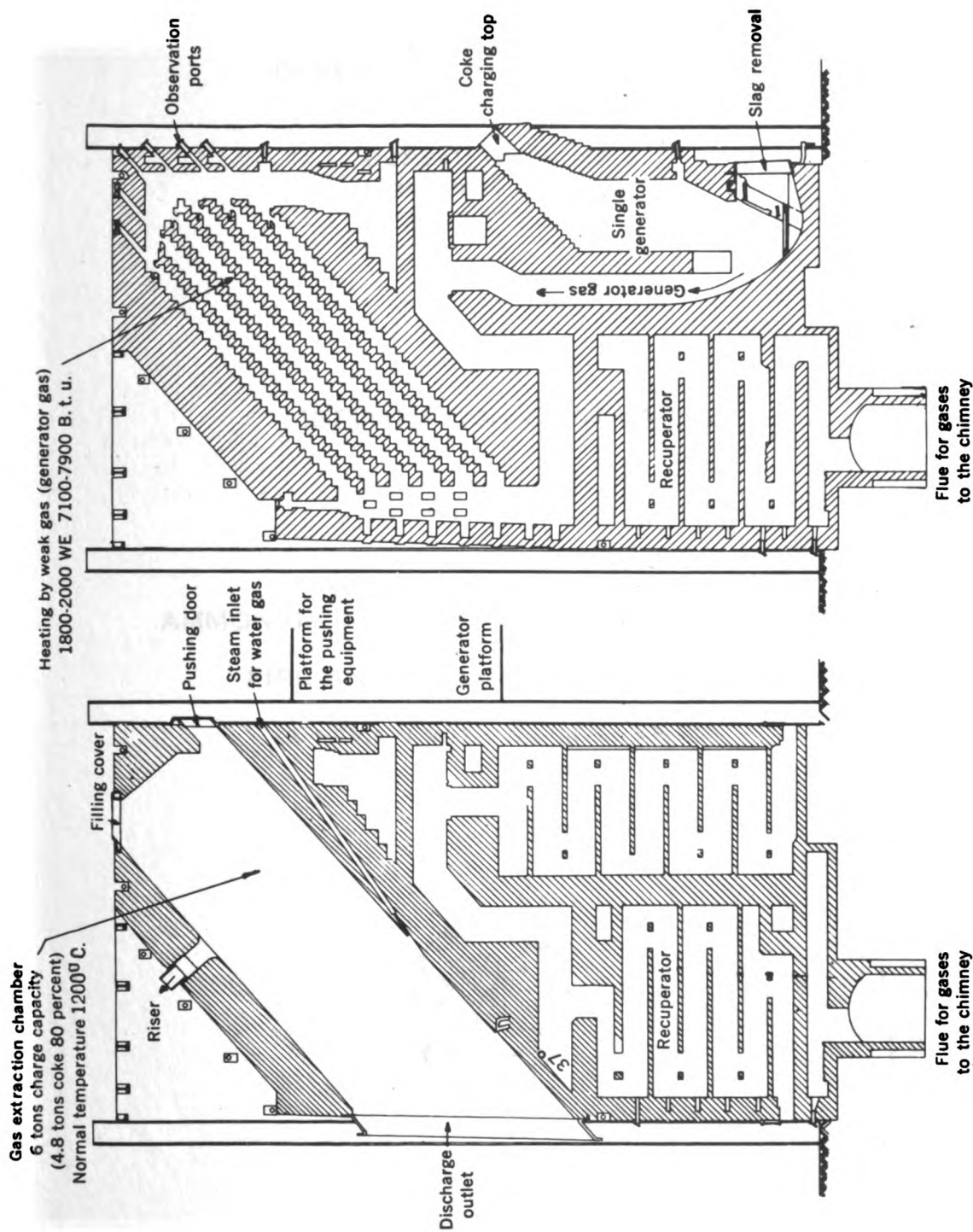


Figure 19. - Inclined chamber oven construction. Didier gas plant, Karlsruhe.